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(54) Title: ELECTROLUMINESCENT BORON COMPLEXES

(57) Abstract: Novel boron complexes are blue emissive electroluminescent compounds.

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## ELECTROLUMINESCENT COMPLEXES

The present invention relates to electroluminescent materials and to electroluminescent devices.

5

Materials which emit light when an electric current is passed through them are well known and used in a wide range of display applications. Liquid crystal devices and devices which are based on inorganic semiconductor systems are widely used. However these suffer from the disadvantages of high energy consumption, high cost  
10 of manufacture, low quantum efficiency and the inability to make flat panel displays.

Organic polymers have been proposed as useful in electroluminescent devices, but it is not possible to obtain pure colours; they are expensive to make and have a relatively low efficiency.

15

Another compound which has been proposed is aluminium quinolate, but this requires dopants to be used to obtain a range of colours and has a relatively low efficiency.

20 Patent application WO98/58037 describes a range of lanthanide and transition metal complexes which can be used in electroluminescent devices which have improved properties and give better results. Patent Applications PCT/GB98/01773, PCT/GB99/03619, PCT/GB99/04030, PCT/GB99/04024, PCT/GB99/04028, PCT/GB00/00268 describe electroluminescent complexes, structures and devices  
25 using rare earth chelates.

US Patent 5128587 discloses an electroluminescent device which consists of an organometallic complex of rare earth elements of the lanthanide series sandwiched between a transparent electrode of high work function and a second electrode of low  
30 work function with a hole conducting layer interposed between the

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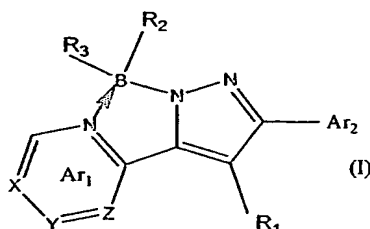
electroluminescent layer and the transparent high work function electrode and an electron conducting layer interposed between the electroluminescent layer and the electron injecting low work function cathode. The hole conducting layer and the electron conducting layer are required to improve the working and the efficiency of the device. The hole transporting layer serves to transport holes and to block the electrons, thus preventing electrons from moving into the electrode without recombining with holes. The recombination of carriers therefore mainly takes place in the emitter layer.

- 10 US Patents 6,287,713 and 6,368,731 the contents of which are incorporated by reference disclose electroluminescent compounds which are complexes of boron with 8-aminoquinolate derivatives.

We have now invented novel electroluminescent boron complexes.

15

According to the invention there is provided a boron compound of formula



wherein:

- Ar<sub>1</sub> represents unsubstituted or substituted monocyclic or polycyclic heteroaryl having a ring nitrogen atom for forming a coordination bond to boron as indicated and optionally one or more additional ring nitrogen atoms subject to the proviso that nitrogen atoms do not occur in adjacent positions, X and Z being carbon or nitrogen and Y being carbon or optionally nitrogen if neither of X and Z is nitrogen, said substituents if present being selected from substituted and unsubstituted hydrocarbyl, substituted and unsubstituted hydrocarbyloxy, fluorocarbon, halo, nitrile, amino alkylamino, dialkylamino or thiophenyl;

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Ar<sub>2</sub> represents monocyclic or polycyclic aryl or heteroaryl optionally substituted with one or more substituents selected from substituted and unsubstituted hydrocarbyl, substituted and unsubstituted hydrocarbyloxy, fluorocarbon, halo, nitrile, amino, alkylamino, dialkylamino or thiophenyl;

5 R<sub>1</sub> represents hydrogen, substituted or unsubstituted hydrocarbyl, halohydrocarbyl or halo; and

R<sub>2</sub> and R<sub>3</sub> each independently represent alkyl, cycloalkyl, cycloalkylalkyl, haloalkyl, halo or monocyclic or polycyclic aryl, heteroaryl, aralkyl or heteroaralkyl optionally substituted with one or more of alkyl, cycloalkyl, cycloalkylalkyl,  
10 haloalkyl, aryl, aralkyl, alkoxy, aryloxy, halo, nitrile, amino, alkylamino or dialkylamino.

Ar<sub>1</sub> typically represents monocyclic or bicyclic heteroaryl in which the ring heteroatoms are nitrogen, e.g. pyridyl, pyrimidyl, pyrazinyl, quinolinyl, iso-  
15 quinolinyl, quinoxalinyl, or quinazolinyl which may be unsubstituted or may be substituted with one or more cycloalkyl, cycloalkylalkyl, haloalkyl, aryl, aralkyl, alkoxy, aryloxy, halo, nitrile, amino, alkylamino or dialkylamino groups. Preferred are groups selected from alkyl, halocarbon or halo substituents e.g. methyl, methoxy, trifluoromethyl or fluoro.

20

Ar<sub>2</sub> typically represents monocyclic or polycyclic aryl, most usually phenyl or naphthyl, but also fluorenyl and 2-6 polycyclic aryl e.g. anthracenyl, phenanthrenyl, pyrenyl or perylenyl which may be unsubstituted or substituted with e.g. alkyl, cycloalkyl, cycloalkylalkyl, haloalkyl, aryl, aralkyl, alkoxy, aryloxy, halo, nitrile,  
25 amino, alkylamino or dialkylamino. Preferred substituents include methyl, methoxy, trifluoromethyl, fluoro and nitrile.

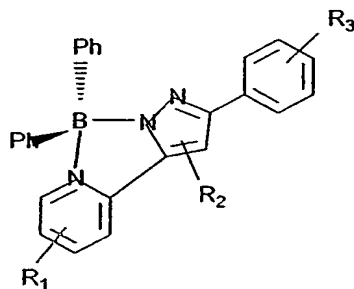
R<sub>1</sub> most usually represents hydrogen, but it may also represent alkyl, cycloalkyl, cycloalkylalkyl, haloalkyl, and monocyclic or polycyclic aryl, heteroaryl, aralkyl and

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heteroaralkyl. Preferred non-hydrogen substituents are methyl, trifluoromethyl and fluoro.

5  $R_2$  and  $R_3$  typically represent phenyl or 4-substituted phenyl wherein the substituent in the 4-position is  $C_1$ - $C_4$  alkyl e.g. methyl or ethyl, trifluoromethyl, methoxy or fluoro. The groups  $R_2$  and  $R_3$  may be derived from e.g. dimethylborinic acid, dimethylborinic anhydride, diethylborinic acid, diethylborinic anhydride, dicyclohexylborinic acid, dicyclohexylborinic anhydride, diphenylborinic acid, diphenylborinic anhydride, , di-p-tolylborinic acid (see US 2002/0161230, Meudt *et al*, Clariant Corporation), and bis(pentafluorophenyl)borinic acid and its anhydride.

According to an alternative definition of the invention there is provided a boron complex of formula



(Ia)

where

Ph is an unsubstituted or substituted phenyl group where the substituents can be the same or different and are selected from hydrogen, and substituted and unsubstituted hydrocarbyl groups such as substituted and unsubstituted aliphatic groups, substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures, fluorocarbons such as trifluoromethyl groups, halogens such as fluorine or thiophenyl groups; and

$R$ ,  $R_1$  and  $R_2$  can be hydrogen or substituted or unsubstituted hydrocarbyl groups, such as substituted and unsubstituted aromatic, heterocyclic and polycyclic

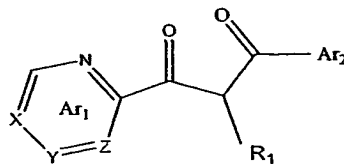
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ring structures, fluorine, fluorocarbons such as trifluoromethyl groups, halogens such as fluorine or thiophenyl groups or nitrile.

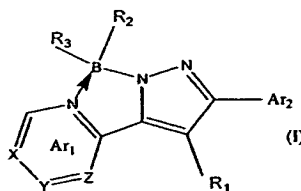
5 Examples of R and/or R<sub>1</sub> and/or R<sub>2</sub> and/or R<sub>3</sub> in Formula (1a) include aliphatic, aromatic and heterocyclic alkoxy, aryloxy and carboxy groups, substituted and substituted phenyl, fluorophenyl, biphenyl, phenanthrene, anthracene, naphthyl and fluorene groups alkyl groups such as t-butyl, heterocyclic groups such as carbazole.

10 The substituents in the above mentioned compounds are advantageously selected so that the compounds sublime without decomposition in vacuo (e.g. at 10<sup>-5</sup> to 10<sup>-7</sup> Torr) at a temperature of from 120-250°C.

15 According to a further aspect of the invention, there is also provided a process for manufacturing a compound of the formula (I) as defined above, which comprises condensing a diketone of the formula:



with hydrazine to give a pyrazole of the formula



20 and esterifying the above pyrazole with a borinic acid of the formula R<sub>2</sub>R<sub>3</sub>BOH or an anhydride of formula R<sub>2</sub>R<sub>3</sub>BOBR<sub>3</sub>R<sub>2</sub> thereof to give the compound of formula (I), atoms X, Y and Z, the rings Ar<sub>1</sub>, Ar<sub>2</sub> and the substituents R<sub>1</sub>-R<sub>3</sub> having the same meanings as for formula (I).

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The starting materials for the diketone may be made by standard methods for the production of 1,3-diones e.g. condensation in the presence of a base of an ester of the corresponding unsubstituted or substituted monocyclic or polycyclic heteroarylcarboxylic acid and the corresponding monocyclic or polycyclic aryl- or heteroaryl-ethanone.

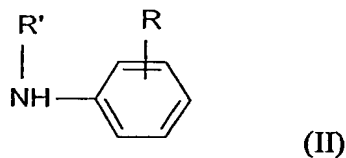
The invention also provides an electroluminescent device which comprises a first electrode, a layer of an electroluminescent material and a second electrode in which the electroluminescent material is a complex of formula (I) or (Ia).

The thickness of the layer of the electroluminescent material is preferably from 10-250 nm, more preferably 20-75 nm.

The first electrode can function as the anode and the second electrode can function as the cathode and preferably there is a layer of a hole transporting material between the anode and the layer of the electroluminescent material.

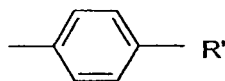
The hole transporting material can be any of the hole transporting materials currently used in electroluminescent devices.

For example, it can be an amine complex such as a-NBP, poly (vinylcarbazole), N, N'-diphenyl-N, N'-bis (3-methylphenyl) -1,1' -biphenyl -4,4'-diamine (TPD), an unsubstituted or substituted polymer of an amino substituted aromatic compound, a polyaniline, substituted polyanilines, polythiophenes, substituted polythiophenes, polysilanes etc. Examples of polyanilines are polymers of



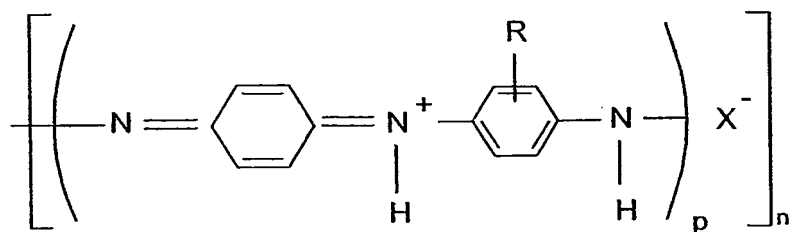
where R is in the ortho – or meta-position and is hydrogen, C1-18 alkyl, C1-6 alkoxy, amino, chloro, bromo, hydroxy or the group

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where R is alkyl or aryl and R' is hydrogen, C1-6 alkyl or aryl with at least one other monomer of formula I above.

- 5 Alternatively the hole transporting material can be a polyaniline. Polyanilines that can be used in the present invention have the general formula



(III)

- where p is from 1 to 10 and n is from 1 to 20, R is as defined above and X is an anion, preferably selected from Cl, Br, SO<sub>4</sub>, BF<sub>4</sub>, PF<sub>6</sub>, H<sub>2</sub>PO<sub>3</sub>, H<sub>2</sub>PO<sub>4</sub>, arylsulphonate, arenedicarboxylate, polystyrenesulphonate, polyacrylate alkylsulphonate, vinylsulphonate, vinylbenzene sulphonate, cellulose sulphonate, camphor sulphonates, cellulose sulphate or a perfluorinated polyanion. Examples of arylsulphonates are p-toluenesulphonate, benzenesulphonate, 9,10-anthraquinone-sulphonate and anthracenesulphonate. An example of an arenedicarboxylate is phthalate and an example of arenecarboxylate is benzoate.
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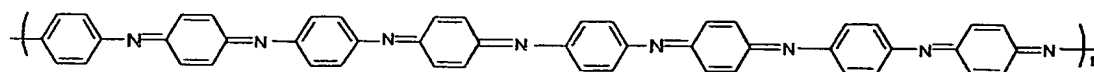
- We have found that protonated polymers of the unsubstituted or substituted polymers of an amino substituted aromatic compound such as a polyaniline are difficult to evaporate or cannot be evaporated. However we have surprisingly found that if the unsubstituted or substituted polymer of an amino substituted aromatic compound is deprotonated then it can be easily evaporated, i.e. the polymer is evaporable. Preferably evaporable deprotonated polymers of unsubstituted or substituted polymers of an amino substituted aromatic compound are used. The de-protonated
- 20



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unsubstituted or substituted polymer of an amino substituted aromatic compound can be formed by deprotonating the polymer by treatment with an alkali such as ammonium hydroxide or an alkali metal hydroxide such as sodium hydroxide or potassium hydroxide. The degree of protonation can be controlled by forming a protonated polyaniline and de-protonating. Methods of preparing polyanilines are described in the article by A. G. MacDiarmid and A. F. Epstein, Faraday Discussions, Chem Soc.88 P319 1989. The conductivity of the polyaniline is dependent on the degree of protonation with the maximum conductivity being when the degree of protonation is between 40 and 60% e.g. about 50% for example. Preferably the polymer is substantially fully deprotonated.

A polyaniline can be formed of octamer units i.e. p is four, e.g.



The polyanilines can have conductivities of the order of  $1 \times 10^{-1}$  Siemen  $\text{cm}^{-1}$  or higher. The aromatic rings can be unsubstituted or substituted e.g. by a  $\text{C}_1$  to  $\text{C}_{20}$  alkyl group such as ethyl.

The polyaniline can be a copolymer of aniline and preferred copolymers are the copolymers of aniline with o-anisidine, m-sulphanilic acid or o-aminophenol, or o-toluidine with o-aminophenol, o-ethylaniline, o-phenylene diamine or with amino anthracenes, o-toluidine, o-ethylaniline, m-toluidine, m-ethylaniline etc.

Other polymers of an amino substituted aromatic compound which can be used include substituted or unsubstituted polyaminonaphthalenes, polyaminoanthracenes, polyaminophenanthrenes, etc. and polymers of any other condensed polyaromatic compound. Polyaminoanthracenes and methods of making them are disclosed in US Patent 6,153,726. The aromatic rings can be unsubstituted or substituted e.g. by a group R as defined above.

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Other hole transporting materials are conjugated polymer and the conjugated polymers which can be used can be any of the conjugated polymers disclosed or referred to in US 5807627, PCT/WO90/13148 and PCT/WO92/03490. The preferred conjugated polymers are poly (p-phenylenevinylene) (PPV) and copolymers including

5 PPV. Other preferred polymers are poly(2,5 dialkoxyphenylene vinylene) such as poly[(2-methoxy-5-(2-methoxypentyloxy-1,4-phenylene vinylene)], poly(2-methoxypentyloxy)-1,4-phenylenevinylene), poly(2-methoxy-5-(2-dodecyloxy-1,4-phenylenevinylene) and other poly(2,5 dialkoxyphenylenevinylenes) with at least one of the alkoxy groups being a long chain solubilising alkoxy group, poly fluorenes and

10 oligofluorenes, polyphenylenes and oligophenylenes, polyanthracenes and oligoanthracenes, polythiophenes and oligothiophenes. In PPV the phenylene ring may optionally carry one or more substituents e.g. each independently selected from alkyl, preferably methyl, alkoxy, preferably methoxy or ethoxy.

15 In poly(fluorene), the fluorene ring may optionally carry one or more substituents e.g. each independently selected from alkyl, preferably methyl, alkoxy, preferably methoxy or ethoxy.

Any poly(arylenevinylene) including substituted derivatives thereof can be used and

20 the phenylene ring in poly(p-phenylenevinylene) may be replaced by a fused ring system such as an anthracene or naphthylene ring and the number of vinylene groups in each polyphenylenevinylene moiety can be increased e.g. up to 7 or higher.

The conjugated polymers can be made by the methods disclosed in US 5807627,

25 PCT/WO90/13148 and PCT/WO92/03490.

The thickness of the hole transporting layer is preferably 20nm to 200nm.

The polymers of an amino substituted aromatic compound such as polyanilines

30 referred to above can also be used as buffer layers with or in conjunction with other

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hole transporting materials e.g. between the anode and the hole transporting layer. Other buffer layers can be formed of phthalocyanines such as copper phthalocyanine.

5 The structural formulae of some other hole transporting materials are shown in Figures 3 to 7 of the drawings, where  $R_1$ ,  $R_2$  and  $R_3$  can be the same or different and are selected from hydrogen, and substituted and unsubstituted hydrocarbyl groups such as substituted and unsubstituted aliphatic groups, substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures, fluorocarbons such as trifluoromethyl groups, halogens such as fluorine or thiophenyl groups;  $R_1$ ,  $R_2$  and  $R_3$  can also form substituted and unsubstituted fused aromatic, heterocyclic and polycyclic ring structures and can be copolymerisable with a monomer e.g. styrene. X is Se, S or O, Y can be hydrogen, substituted or unsubstituted hydrocarbyl groups, such as substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures, fluorine, fluorocarbons such as trifluoromethyl groups, halogens such as fluorine or thiophenyl groups or nitrile.

Examples of  $R_1$  and/or  $R_2$  and/or  $R_3$  include aliphatic, aromatic and heterocyclic alkoxy, aryloxy and carboxy groups, substituted and substituted phenyl, fluorophenyl, biphenyl, phenanthrene, anthracene, naphthyl and fluorene groups alkyl groups such as t-butyl, heterocyclic groups such as carbazole.

Optionally there is a layer of an electron injecting material between the cathode and the electroluminescent material layer. The electron injecting material is a material which will transport electrons when an electric current is passed through electron injecting materials including TAZ (formula in Fig. 2) or a metal complex such as a metal quinolate e.g. a zirconium quinolate ( $Zr_q4$ ), zinc quinolate, aluminium quinolate, lithium quinolate,  $M_x(DBM)_n$  where  $M_x$  is a metal and DBM is dibenzoyl methane and n is the valency of  $M_x$  e.g.  $M_x$  is aluminium or chromium.

In place of the DBM moiety there can be a Schiff base.

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The electron injecting material can also be a cyano anthracene such as 9,10 dicyano anthracene, cyano substituted aromatic compounds, tetracyanoquinodimethane a polystyrene sulphonate or a compound with the structural formulae shown in figures 1 or 2 of the drawings in which the phenyl rings can be substituted with substituents R as defined above. Instead of being a separate layer the electron injecting material can be mixed with the electroluminescent material and co-deposited with it.

Optionally the hole transporting material can be mixed with the electroluminescent material and co-deposited with it and the electron injecting materials and electroluminescent materials can be mixed. The hole transporting materials, the electroluminescent material and the electron injecting materials can be mixed together to form one layer, which simplifies the construction.

The anode is preferably a transparent substrate such as a conductive glass or plastic material which acts as the anode. Preferred substrates are conductive glasses such as indium tin oxide coated glass, but any glass which is conductive or has a conductive layer such as a metal or conductive polymer can be used. Conductive polymers and conductive polymer coated glass or plastics materials can also be used as the substrate. The cathode is preferably a low work function metal e.g. aluminium, barium, rare earth metals, transition metals, calcium, lithium, magnesium and alloys thereof such as silver/magnesium alloys, rare earth metal alloys etc.; aluminium is a preferred metal. A metal fluoride such as an alkali metal e.g. lithium fluoride or rare earth metal fluoride can be used as the second electrode for example by having a metal fluoride layer formed on a metal.

The boron complexes of the present invention include blue emitting electroluminescent materials.

The devices of the present invention can be used as displays in video displays, mobile telephones, portable computers and any other application where a electronically

controlled visual image is used. The devices of the present invention can be used in both active and passive applications of such as displays.

5 In known electroluminescent devices either one or both electrodes can be formed of silicon and the electroluminescent material and intervening layers of a hole transporting and electron transporting materials can be formed as pixels on the silicon substrate. Preferably each pixel comprises at least one layer of an electroluminescent material and a (at least semi-) transparent electrode in contact with the organic layer on a side thereof remote from the substrate.

10

Preferably, the substrate is of crystalline silicon and the surface of the substrate may be polished or smoothed to produce a flat surface prior to the deposition of electrode, or electroluminescent compound. Alternatively a non-planarised silicon substrate can be coated with a layer of conducting polymer to provide a smooth, flat surface prior to deposition of further materials.

15

In one embodiment, each pixel comprises a metal electrode in contact with the substrate. Depending on the relative work functions of the metal and transparent electrodes, either may serve as the anode with the other constituting the cathode.

20

When the silicon substrate is the cathode an indium tin oxide coated glass can act as the anode and light is emitted through the anode. When the silicon substrate acts as the anode, the cathode can be formed of a transparent electrode which has a suitable work function; for example by a indium zinc oxide coated glass in which the indium zinc oxide has a low work function. The anode can have a transparent coating of a metal formed on it to give a suitable work function. These devices are sometimes referred to as top emitting devices or back emitting devices.

25

The metal electrode may consist of a plurality of metal layers; for example a higher work function metal such as aluminium deposited on the substrate and a lower work

30

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function metal such as calcium deposited on the higher work function metal. In another example, a further layer of conducting polymer lies on top of a stable metal such as aluminium.

- 5 Preferably, the electrode also acts as a mirror behind each pixel and is either deposited on, or sunk into, the planarised surface of the substrate. However, there may alternatively be a light absorbing black layer adjacent to the substrate.

- 10 In still another embodiment, selective regions of a bottom conducting polymer layer are made non-conducting by exposure to a suitable aqueous solution allowing formation of arrays of conducting pixel pads which serve as the bottom contacts of the pixel electrodes.

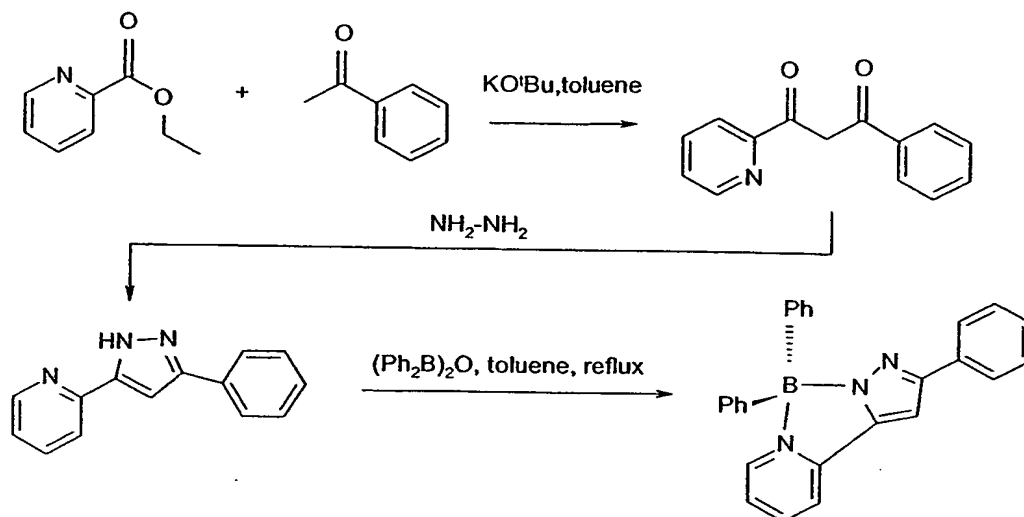
- 15 Blue electroluminescent materials are the most difficult to obtain with satisfactory performance and, in blue monochromatic displays and polychromatic displays, a blue electroluminescent material of the present invention can be used.

- 20 As described in WO00/60669 the brightness of light emitted from each pixel is preferably controllable in an analogue manner by adjusting the voltage or current applied by the matrix circuitry or by inputting a digital signal which is converted to an analogue signal in each pixel circuit. The substrate preferably also provides data drivers, data converters and scan drivers for processing information to address the array of pixels so as to create images.

- 25 In a further embodiment, each pixel is controlled by a switch comprising a voltage controlled element and a variable resistance element, both of which are conveniently formed by metal-oxide-semiconductor field effect transistors (MOSFETs) or by an active matrix transistor.

- 30 The invention is illustrated in the following examples

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**Example 1***Preparation of 2-(2-diphenylboranyl-5-phenylpyrazol-3-yl)pyridine*

- 5           Acetophenone (11.6 mL, 99 mmol), was added to a suspension of potassium tert-butoxide (12.34g 110 mmol) in toluene (150 mL) and stirred under argon. The flask was then charged with ethyl picolinate (13.4 mL, 99 mmol) and the mixture stirred overnight. The toluene was removed by rotary evaporation and 200 mL of both diethylether and deionised water added. With stirring the mixture was acidified with
- 10   dilute HCl until the ether layer was dark orange ~ pH 7). The ether layer was separated, washed with deionised water (3 x 100 mL), dried over magnesium sulfate and filtered. The solvent was removed and the solid recrystallised from hot ethanol to yield 1-phenyl-3-pyridin-2-ylpropane-1,3-dione (1) as a pale yellow crystalline solid (15.3 g, 69%).

15

- A 250 mL flask was charged with compound (1) (10g 44 mmol), a magnetic follower and 150 mL of ethanol. Hydrazine monohydrate (2.15 mL, 44 mmol) was added and the mixture refluxed overnight. On cooling the ethanol was removed and the residue recrystallised from methanol to yield 2-(5-phenylpyrazol-3-yl)pyridine (2)
- 20   as a pale yellow crystalline solid (9.5 g, 97 %).

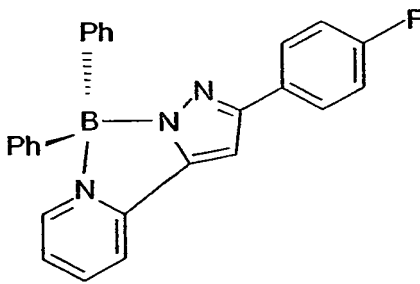
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A 150 mL flask was charged with diphenylborinic anhydride (1.00 g, 2.9 mmol), compound (2) (1.28 g, 5.8 mmol) and 100 mL of toluene. The mixture was refluxed until all traces of the anhydride had dissolved and then refluxed for a further 2 hours. On cooling, white crystals were formed. These were collected by filtration and dried under vacuum to give 2.15g (96%) of 2-(2-diphenylboranyl-5-phenylpyrazol-3-yl)pyridine. DSC:- Mpt. 261.4- 265.2° C (Recryst. 173.3 °C). The properties of this complex are shown in table 1 below and its emission spectrum and UV absorbance are shown in figs. 8 and 9.

10

**Example 2**

*Preparation of 2-(2-diphenylboranyl-5-(4-fluorophenyl)pyrazol-3-yl)pyridine*



2-(2-diphenylboranyl-5-(4-fluorophenyl)pyrazol-3-yl)pyridine was prepared by the method of example 1 with acetophenone being replaced by 4'-fluoroacetophenone.

The diketone was prepared in ~70% yield and the free ligand and boron compounds were formed in 90+% yields. DSC;\_Mpt. 263.7- 266.6°C (Recryst.196.4°C). The properties of this complex are shown in table 1 below and its emission spectrum and UV absorbance are shown in figs. 10 and 11.

20

**Table 1**

Compound of Example	PL Efficiency Cdm <sup>-2</sup> μw <sup>-1</sup>	Peak Wavelength nm	CIE Coord. x;y	M.P.°C
1	0.071	~455	0.17;0.16	263-267
2	0.058	~450	0.16;0.15	261-265

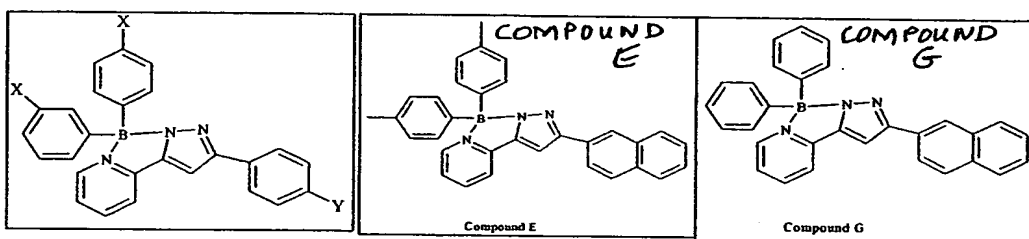
The CIE coordinates refer to the CIE xy chromaticity diagram.



## Examples 3 – 12

The compounds indicated in Table II were prepared as in Example 1, and their properties were as shown in the Table, in which the x and y values in the final two columns are colour coordinates.

Table II - Compounds of Examples 3-12



Identity (Ex.)	X	Y	DSC m.p. (°C)	DSC Tg (°C)	PL (Film) Peak (nm)	PL (Film) Efficiency (cdm <sup>-2</sup> μW <sup>-1</sup> )	PL (Film) x	PL (Film) y
A(3)	H	H	259.1-263.1	Not observed	460	0.111	0.1593	0.1968
C(4)	H	CN	328.2-334.4	Not observed	460	0.043	0.1543	0.1257
D(5)	Me	H	251.8 - 256	95.2	460	0.036	0.1626	0.1822
E(6)	Figure E		287-291	116 Not	475	0.09	0.1714	0.2578
F(7)	H	F	383 - 393	observed	461	0.112	0.1619	0.2015
G(8)	Figure G		313.6-317.7	106.8	470	0.09	0.1706	0.2572
H(9)	H	Me	267.5-271.9	97.6	465	0.06	0.1668	0.2261
I(10)	OMe	Me	295-300	95	468	0.087	0.1740	0.2489
K(11)	Me	F	244-248	100	460	0.06	0.1679	0.1858
L(12)	Me	CN	322.6-327.6	116.9	440	0.03	0.1625	0.1822

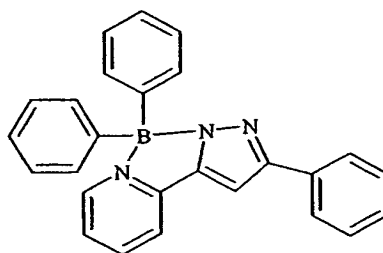
- 17 -

**Example 13**

A pre-etched ITO (indium tin oxide) coated glass piece (10 x 10cm<sup>2</sup> ) was used. A device was fabricated by sequentially forming layers on the ITO by vacuum evaporation, using a Solciet Machine, ULVAC Ltd. Chigacki, Japan; the active area of each pixel was 3mm by 3mm. The layers comprised:-

ITO (H)/CuPc (25 nm)/ $\alpha$ -NPB (110 nm)/Compound A (35 nm)/Al

10 in which compound A has the formula



Compound A

CuPc is a copper phthalocyanine buffer layer and  $\alpha$ -NPB is as in fig. 7. The coated electrode was stored in a vacuum desiccator over a molecular sieve and phosphorous pentoxide until it was loaded into a vacuum coater (10<sup>-6</sup> torr) and the aluminium top contact was made. The device was then kept in a vacuum desiccator until the electroluminescence studies were performed.

In these studies, the ITO electrode was always connected to the positive terminal. The current vs. voltage studies were carried out on a computer controlled Keithly 2400 source meter. The electroluminescent spectrum was measured and the results shown in Fig. 12. Luminescence and current density were measured as a function of voltage and luminescence and current efficiency were measured as a function of current density with the results shown in Fig. 13.

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A further electroluminescent device was made with a cell comprising the following layers, LiF representing lithium fluoride:

ITO (H)/CuPc (25 nm)/ $\alpha$ -NPB (85 nm)/Compound A (55 nm)/LiF (0.2 nm)/Al.

5

The electroluminescent spectrum was measured and the results shown in Fig. 14.

Luminescence and current density were measured as a function of voltage and

luminescence and current efficiency were measured as a function of current density

with the results shown in Fig. 15. A third electroluminescent device was made with a

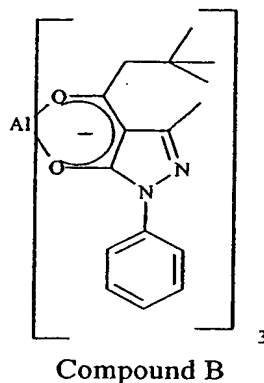
10 cell comprising the following layers:

ITO(H)/CuPc (25 nm)/ $\alpha$ -NPB (110 nm)/Compound A (40 nm)/Zr<sub>q</sub><sub>4</sub> (10 nm)/LiF (0.2 nm)/Al

15 in which ZrQ<sub>4</sub> is zirconium quinolate. The electroluminescent spectrum was measured and the results are shown in Fig. 16. Luminescence and current density were measured as a function of voltage and luminescence and current efficiency were measured as a function of current density with the results shown in Fig. 17. A fourth

20

ITO(H)/CuPc (25 nm)/ $\alpha$ -NPB (110 nm)/Compound B (1 nm)/Compound A (40 nm)/Zr<sub>q</sub><sub>4</sub> (10 nm)/LiF (0.2 nm)/Al in which compound B is of formula:



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The electroluminescent spectrum was measured and the results are shown in Fig. 18. Luminescence and current density were measured as a function of voltage and luminescence and current efficiency were measured as a function of current density with the results shown in Fig. 19.

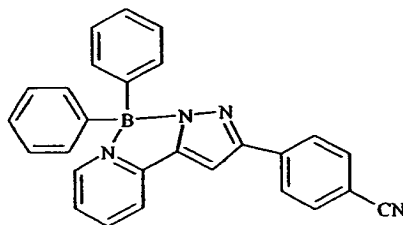
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#### Example 14

An electroluminescent device was made with the following layers:

10 ITO(H)/CuPc (25 nm)/ $\alpha$ -NPB (110 nm)/Compound C (40 nm)/Zr<sub>q</sub>4 (10 nm)/LiF (0.5 nm)/Al

in which compound C is of formula:



Compound C

15 The electroluminescent spectrum and other properties were measured and the results are shown in Figures 20 and 21.

A second electroluminescent device was made with the following layers:

20 ITO(H)/CuPc (25 nm)/ $\alpha$ -NPB (75 nm)/BAI<sub>q</sub>2 (15 nm)/Compound C (40 nm)/Zr<sub>q</sub>4 (10 nm)/LiF (0.5 nm)/Al

In the above structure BAI<sub>q</sub>2 represents biphenyl aluminium biquinolate (the same as the compound BAI<sub>q</sub>1 in Fig 1). The electroluminescent spectrum was measured and  
25 the results are shown in Fig. 22. Luminescence and current density were measured as

- 20 -

a function of voltage and luminescence and current efficiency were measured as a function of current density with the results shown in Fig. 23.

### Example 15

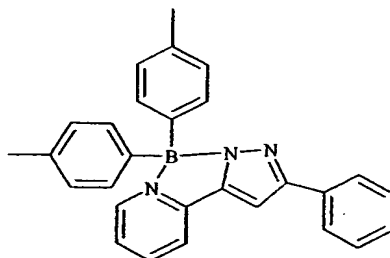
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An electroluminescent device was made with the following layers:

ITO(H)/CuPc (25 nm)/ $\alpha$ -NPB (110 nm)/Compound D (40 nm)/BAIq<sub>2</sub> (10 nm)/LiF (0.5 nm)/Al

10

in which compound D is of formula:



Compound D

The electroluminescent spectrum was measured and the results are shown in Fig. 24. Luminescence and current density were measured as a function of voltage and luminescence and current efficiency were measured as a function of current density with the results shown in Fig. 25.

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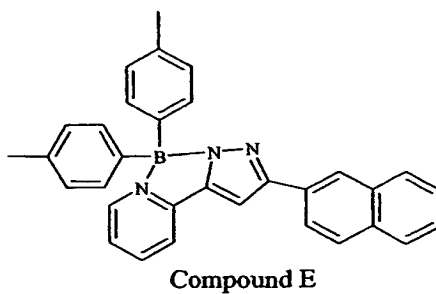
### Example 16

20 An electroluminescent device was made with the following layers:

ITO(H)/CuPc (25 nm)/ $\alpha$ -NPB (60 nm)/Compound E:Perylene (30:0.02 nm)/BAIq<sub>2</sub> (10 nm)/LiF (0.5 nm)/Al

- 21 -

in which compound E is of formula:



The electroluminescent spectrum was measured and the results are shown in Fig. 26.

Luminescence and current density were measured as a function of voltage and

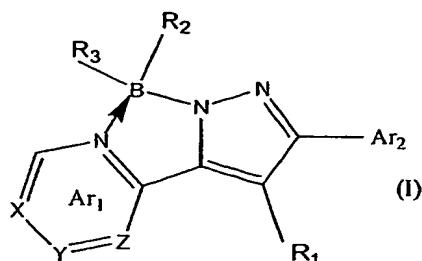
5 luminescence and current efficiency were measured as a function of current density  
with the results shown in Fig. 27.

10

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## CLAIMS

1. A boron compound of formula



- 5 wherein:

Ar<sub>1</sub> represents unsubstituted or substituted monocyclic or polycyclic heteroaryl having a ring nitrogen atom for forming a coordination bond to boron as indicated and optionally one or more additional ring nitrogen atoms subject to the proviso that nitrogen atoms do not occur in adjacent positions, X and Z being carbon or nitrogen and Y being carbon or optionally nitrogen if neither of X and Z is nitrogen, said substituents if present being selected from substituted and unsubstituted hydrocarbyl, substituted and unsubstituted hydrocarbyloxy, fluorocarbon, halo, nitrile, amino alkylamino, dialkylamino or thiophenyl;

Ar<sub>2</sub> represents monocyclic or polycyclic aryl or heteroaryl optionally substituted with one or more substituents selected from substituted and unsubstituted hydrocarbyl, substituted and unsubstituted hydrocarbyloxy, fluorocarbon, halo, nitrile, amino, alkylamino, dialkylamino or thiophenyl;

R<sub>1</sub> represents hydrogen, substituted or unsubstituted hydrocarbyl, haloalkyl or halo; and

R<sub>2</sub> and R<sub>3</sub> each independently represent alkyl, cycloalkyl, cycloalkylalkyl, haloalkyl, halo or monocyclic or polycyclic aryl, heteroaryl, aralkyl or heteroaralkyl optionally substituted with one or more of alkyl, cycloalkyl, cycloalkylalkyl, haloalkyl, aryl, aralkyl, alkoxy, aryloxy, halo, nitrile, amino, alkylamino or dialkylamino.

25

- 23 -

2. The compound of claim 1, wherein Ar<sub>1</sub> represents monocyclic or bicyclic heteroaryl which is unsubstituted or is substituted with one or more of C<sub>1</sub>-C<sub>4</sub> alkyl C<sub>1</sub>-C<sub>4</sub> alkoxy, trifluoromethyl or fluoro.
- 5 3. The compound of claim 2, wherein Ar<sub>1</sub> represents monocyclic or bicyclic heteroaryl which is unsubstituted or substituted pyridyl, pyrimidyl, pyrazinyl, quinolinyl, iso-quinolinyl, quinoxaliny, or quinazolinyl..
4. The compound of any preceding claim, wherein Ar<sub>2</sub> represents monocyclic or  
10 2-6 polycyclic aryl which is unsubstituted.
5. The compound of claim 4, wherein the monocyclic or polycyclic aryl is substituted with one or more of C<sub>1</sub>-C<sub>4</sub> alkyl, C<sub>1</sub>-C<sub>4</sub> alkoxy, trifluoromethyl, fluoro or nitrile.
- 15 6. The compound of claim 4 or 6, wherein the monocyclic or polycyclic aryl is phenyl, naphthyl, anthracenyl, phenanthrenyl, pyrenyl or peryleneyl.
7. The compound of any preceding claim, wherein R<sub>1</sub> represents hydrogen.
- 20 8. The compound of any of claims 1-6, wherein R<sub>1</sub> represents alkyl, cycloalkyl, cycloalkylalkyl, haloalkyl, monocyclic or polycyclic aryl, heteroaryl, aralkyl or heteroaralkyl.
- 25 9. The compound of any of claims 1-6, wherein R<sub>1</sub> represents C<sub>1</sub>-C<sub>4</sub> alkyl, trifluoromethyl or fluoro.
10. The compound of any preceding claim, wherein R<sub>2</sub> and R<sub>3</sub> represent phenyl or 4-substituted phenyl.

30



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11. The compound of claim 10, wherein the substituent in the 4-position is C<sub>1</sub>-C<sub>4</sub> alkyl, trifluoromethyl, C<sub>1</sub>-C<sub>4</sub> alkoxy or fluoro.
12. An electroluminescent device, which comprises a first electrode, a layer of an electroluminescent material as claimed in any preceding claim and a second electrode.
13. The device of claim 12, wherein there is a layer of a hole transmitting material between the first electrode, which functions as the anode, and the layer of the electroluminescent material.
14. The device of claim 13, wherein which the hole transmitting material is an aromatic amine complex.
15. The device of claim 13, wherein the hole transmitting material is a polyaromatic amine complex.
16. The device of claim 13, wherein the hole transmitting material is a film of a polymer selected from a-NPB, poly(vinylcarbazole), N,N'-diphenyl-N,N'-bis (3-methylphenyl) -1,1' -biphenyl -4,4'-diamine (TPD), CBP, polyaniline, substituted polyanilines, polythiophenes, substituted polythiophenes, polysilanes and substituted polysilanes.
17. The device of claim 13, wherein the hole transmitting material is a film of a compound of formula (II) or (III) herein or as in figures 3 to 7 of the drawings.
18. The device of claim 13, wherein the hole transmitting material is a copolymer of aniline, a copolymer of aniline with o-anisidine, m-sulphanilic acid or o-aminophenol, or o-toluidine with o-aminophenol, o-ethylaniline, o-phenylene diamine or with an amino anthracene, o-toluidine, o-ethylaniline, m-toluidine, m-ethylaniline.

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19. The device of claim 13, wherein the hole transmitting material is a conjugated polymer.

5 20. The device of claim 19, wherein the conjugated polymer is selected from poly (p-phenylenevinylene)-PPV and copolymers including PPV, poly(2,5 dialkoxyphenylene vinylene), poly (2-methoxy-5-(2-methoxypentyloxy-1,4-phenylene vinylene), poly(2-methoxypentyloxy)-1,4-phenylenevinylene), poly(2-methoxy-5-(2-dodecyloxy-1,4-phenylenevinylene) and other poly(2,5  
10 dialkoxyphenylenevinylenes) with at least one of the alkoxy groups being a long chain solubilising alkoxy group, polyfluorenes and oligofluorenes, polyphenylenes and oligophenylenes, polyanthracenes and oligo anthracenes, ploythiophenes and oligothiophenes and substituted polyfluorenes.

15 21. The device of any of claims 12-20, wherein the electroluminescent compound is in admixture with the hole transmitting material.

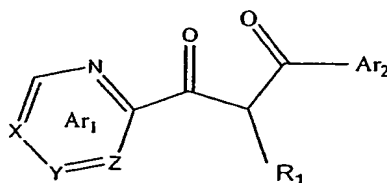
22. The device of any of claims 12-21, wherein there is a layer of an electron transmitting material between the second electrode, which functions as the cathode,  
20 and the layer of the electroluminescent material.

23. The device of claim 22, wherein the electron transmitting material is a metal quinolate.

25 24. The device of claim 23, wherein the metal quinolate is an aluminium quinolate or lithium quinolate.

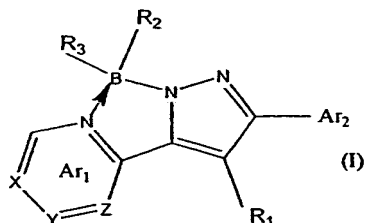
- 26 -

25. The device of claim 22, wherein the electron transmitting material is of formula  $Mx(DBM)_n$  where  $Mx$  is a metal and DBM is dibenzoyl methane and  $n$  is the valency of  $Mx$  or there is a Schiff base in place of the DBM.
- 5 26. The device of claim 22, wherein the electron transmitting material is a cyano anthracene, 9,10 dicyano anthracene, a polystyrene sulphonate or a compound of formulae shown in figure 1 or 2 of the drawings.
- 10 27. The device of any of claims 22-26, wherein the electron transmitting material is mixed with the electroluminescent compound.
28. The device of any of claims 12-27, wherein the first electrode is a transparent electricity conducting glass electrode.
- 15 29. The device of any of claims 12-28, wherein the second electrode is selected from aluminium, barium, rare earth metals, transition metals, calcium, lithium, magnesium and alloys thereof and silver/magnesium alloys.
- 20 30. The device of any of claims 12-29, wherein the second electrode is selected from a metal having a metal fluoride layer formed on it.
31. The device of claim 30, wherein the metal fluoride is a lithium fluoride or rare earth fluoride.
- 25 32. A process for manufacturing a compound of the formula (I) as defined in claim 1, which comprises condensing a diketone of the formula:



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with hydrazine to give a pyrazole of the formula

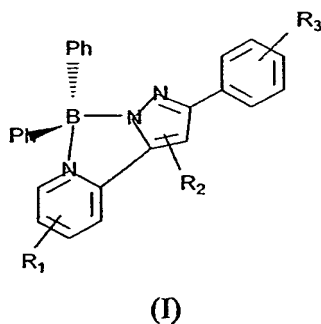


and esterifying the above pyrazole with a borinic acid of the formula  $R_2R_3BOH$  or an anhydride of formula  $R_2R_3BOBR_3R_2$  thereof to give the compound of formula (I),  
 5 atoms X, Y and Z, the rings  $Ar_1$ ,  $Ar_2$  and the substituents  $R_1$ - $R_3$  having the same meanings as in claim 1.

33. The process of claim 32, further comprising the step of making the diketone  
 10 of claim 32 by condensing in the presence of a base an ester of the an unsubstituted or substituted monocyclic or polycyclic heteroarylcarboxylic acid with a monocyclic or polycyclic aryl- or heteroaryl-ethanone.

34. The process of claim 32 or 33, which comprises esterifying the pyrazole with  
 15 dimethylborinic acid, dimethylborinic anhydride, diethylborinic acid, diethylborinic anhydride, dicyclohexylborinic acid, dicyclohexylborinic anhydride, diphenylborinic acid, diphenylborinic anhydride, di-p-tolylborinic acid, bis(pentafluorophenyl)borinic acid or its anhydride.

35. A boron complex of formula  
 20



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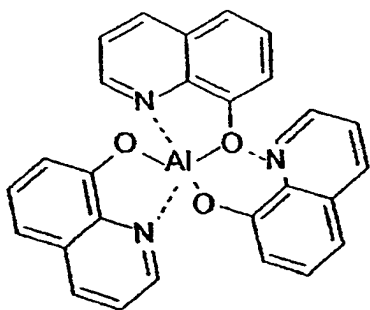
where

Ph is an unsubstituted or substituted phenyl group where the substituents can be the same or different and are selected from hydrogen, and substituted and unsubstituted hydrocarbyl groups such as substituted and unsubstituted aliphatic groups, substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures, fluorocarbons such as trifluoromethyl groups, halogens such as fluorine or thiophenyl groups; and

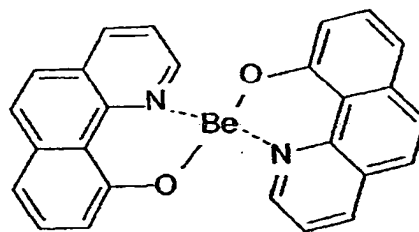
R, R<sub>1</sub> and R<sub>2</sub> can be hydrogen or substituted or unsubstituted hydrocarbyl groups, such as substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures, fluorine, fluorocarbons such as trifluoromethyl groups, halogens such as fluorine or thiophenyl groups or nitrile.

36. A complex as claimed in claim 35, in which R and/or R<sub>1</sub> and/or R<sub>2</sub> are aliphatic, aromatic or heterocyclic alkoxy, aryloxy and carboxy groups, substituted and substituted phenyl, fluorophenyl, biphenyl, phenanthrene, anthracene, naphthyl and fluorene groups alkyl groups such as t-butyl, heterocyclic groups such as carbazole.

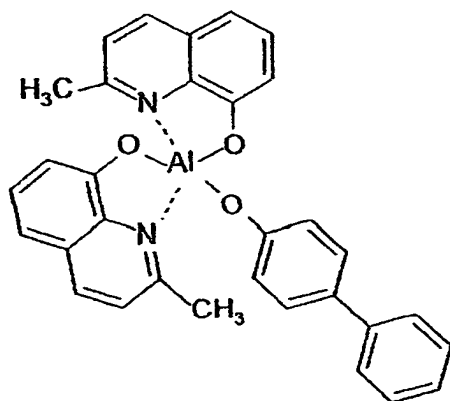
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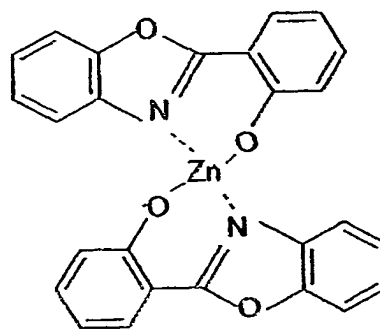
Alq



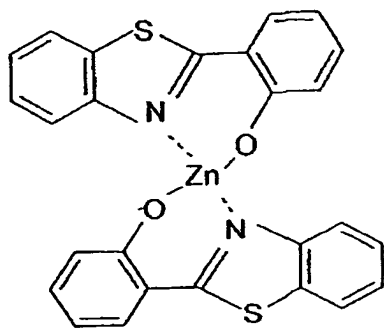
Bebq



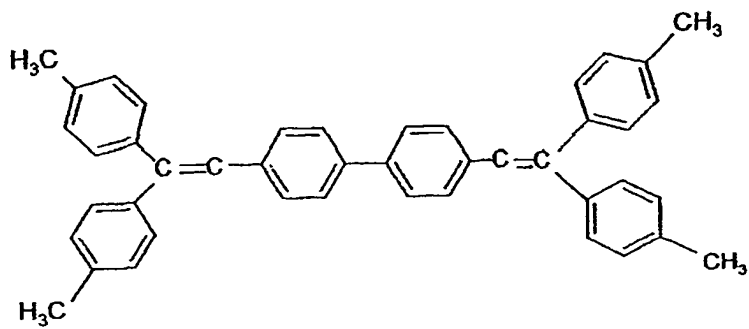
BAlq1



ZnPBO



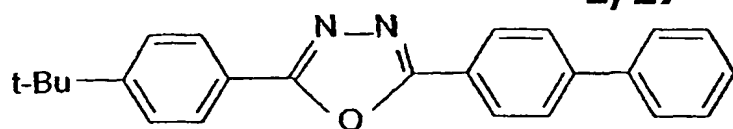
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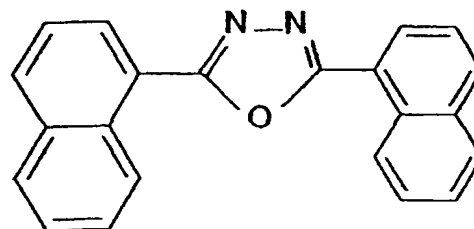
DTVb1

Fig 1

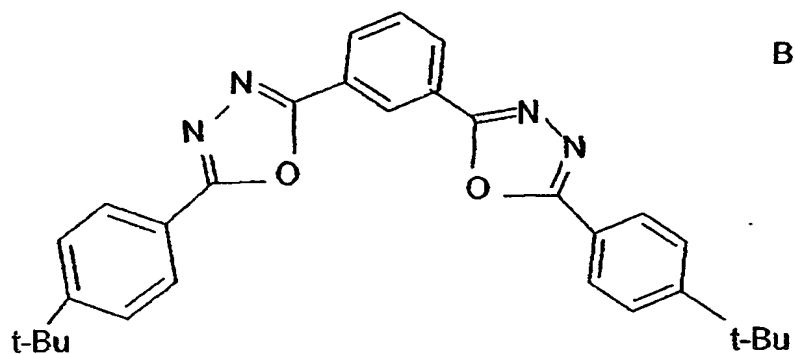
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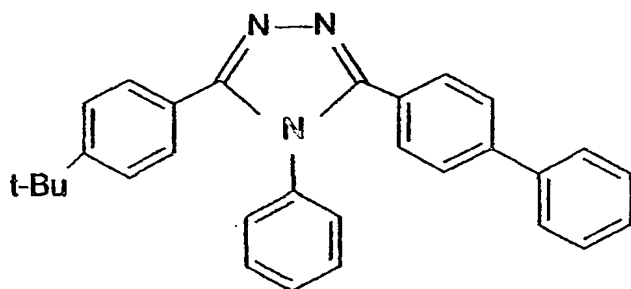
t-Bu-PBD



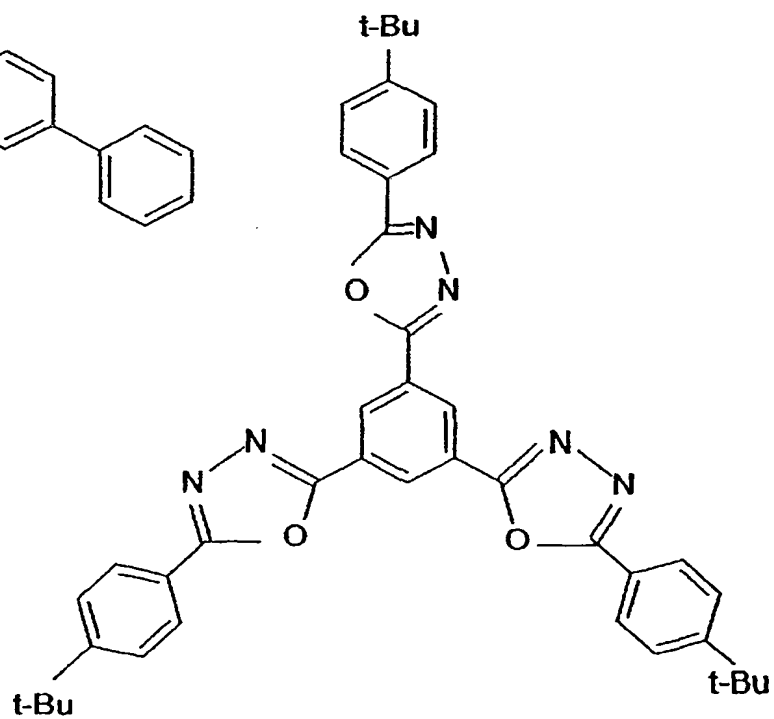
BND



OXD-7



TAZ



OXD-Star

Fig 2

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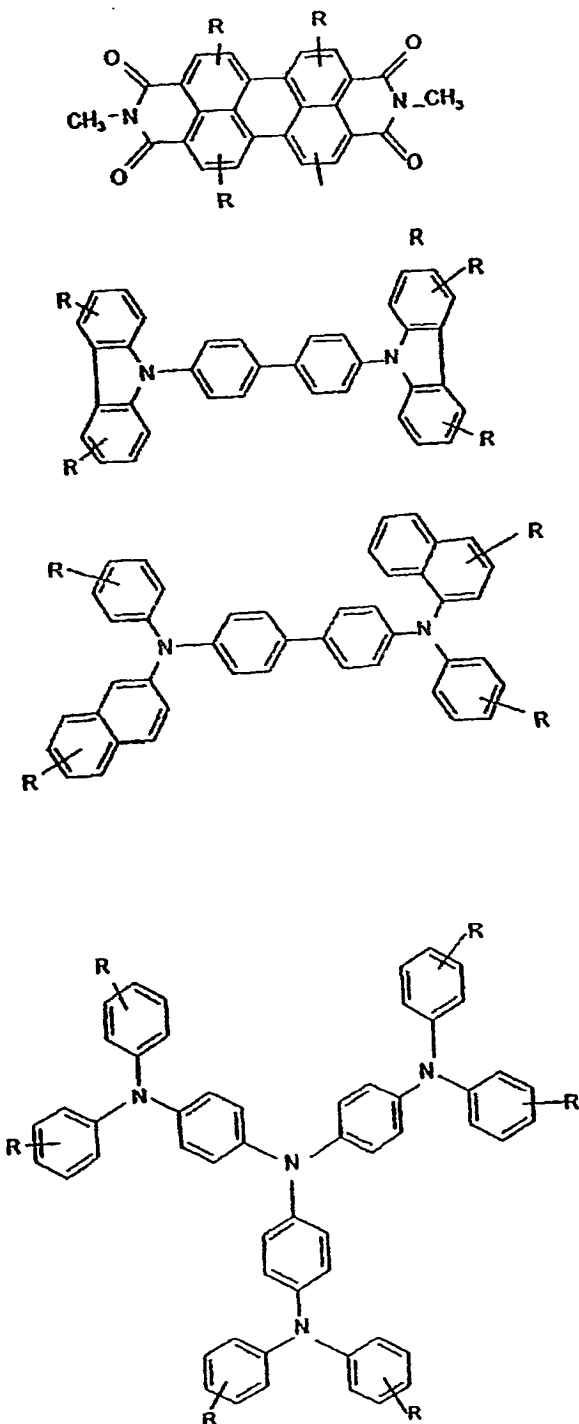


Fig 3



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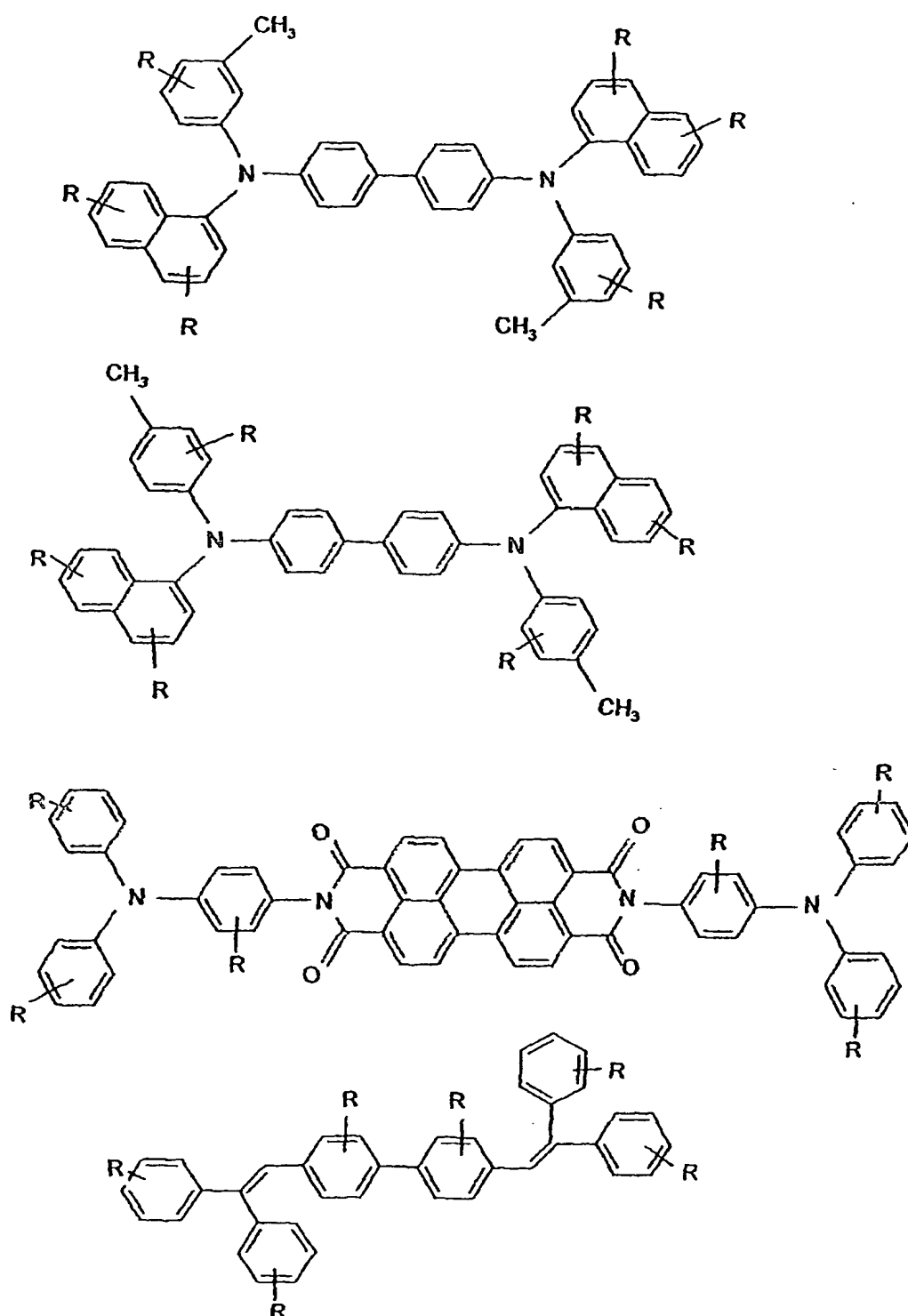


Fig 4

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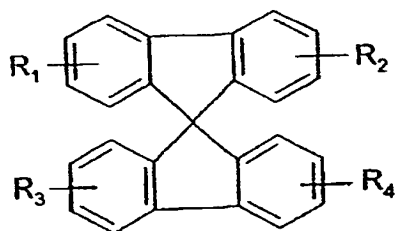


Fig. 14a

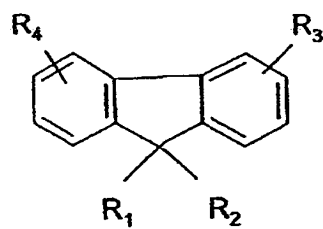
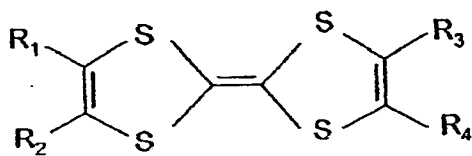


Fig. 14b



or

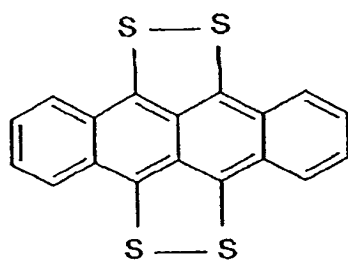
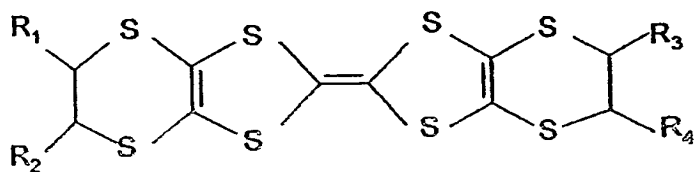


Fig 5

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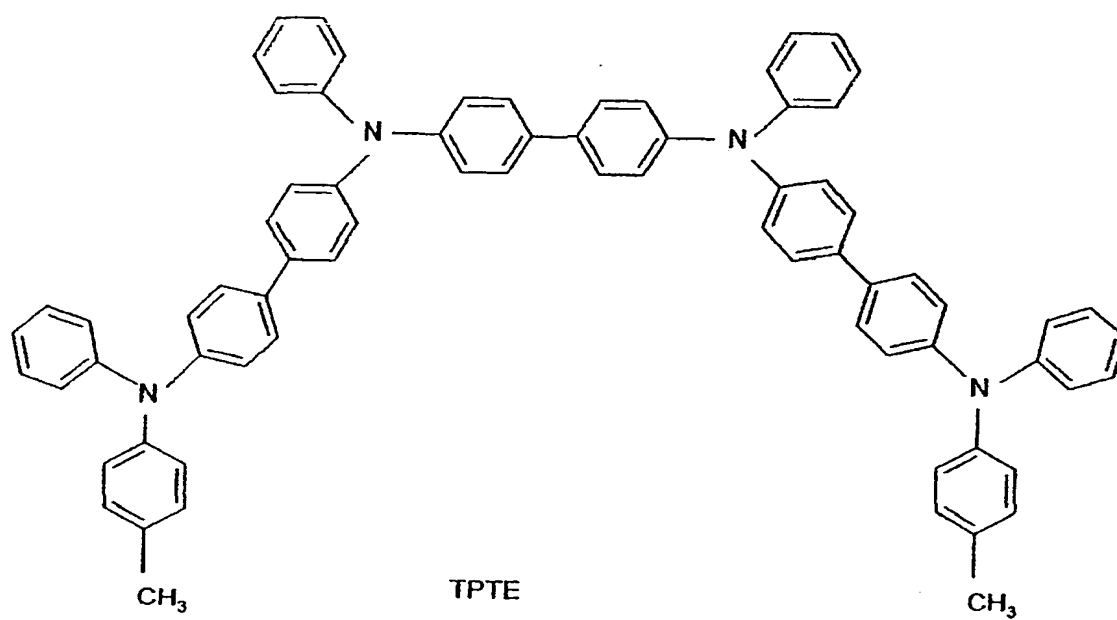
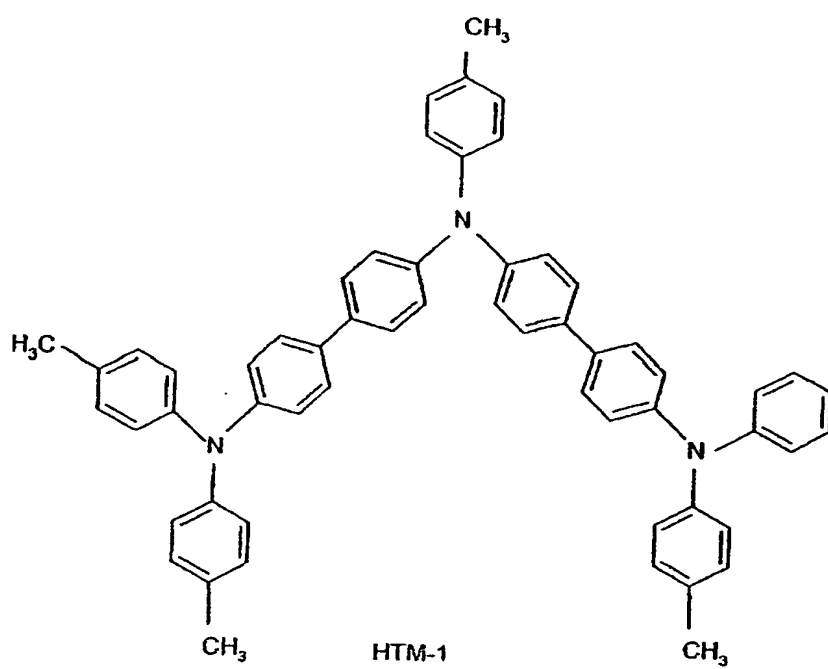


Fig 6

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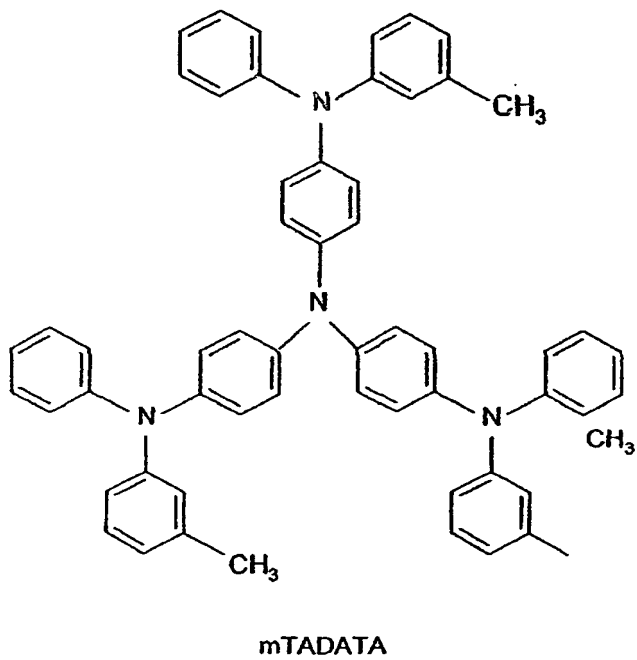
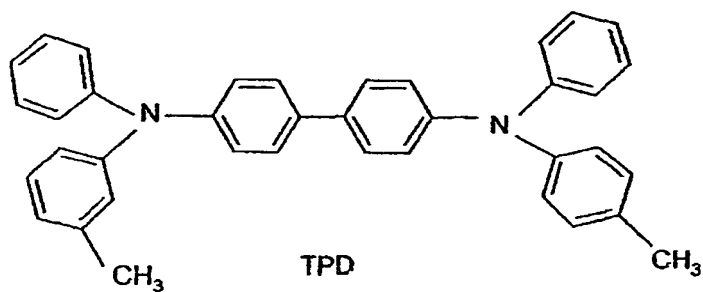
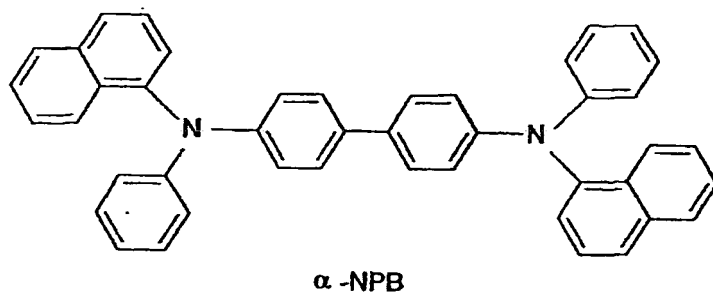


Fig 7

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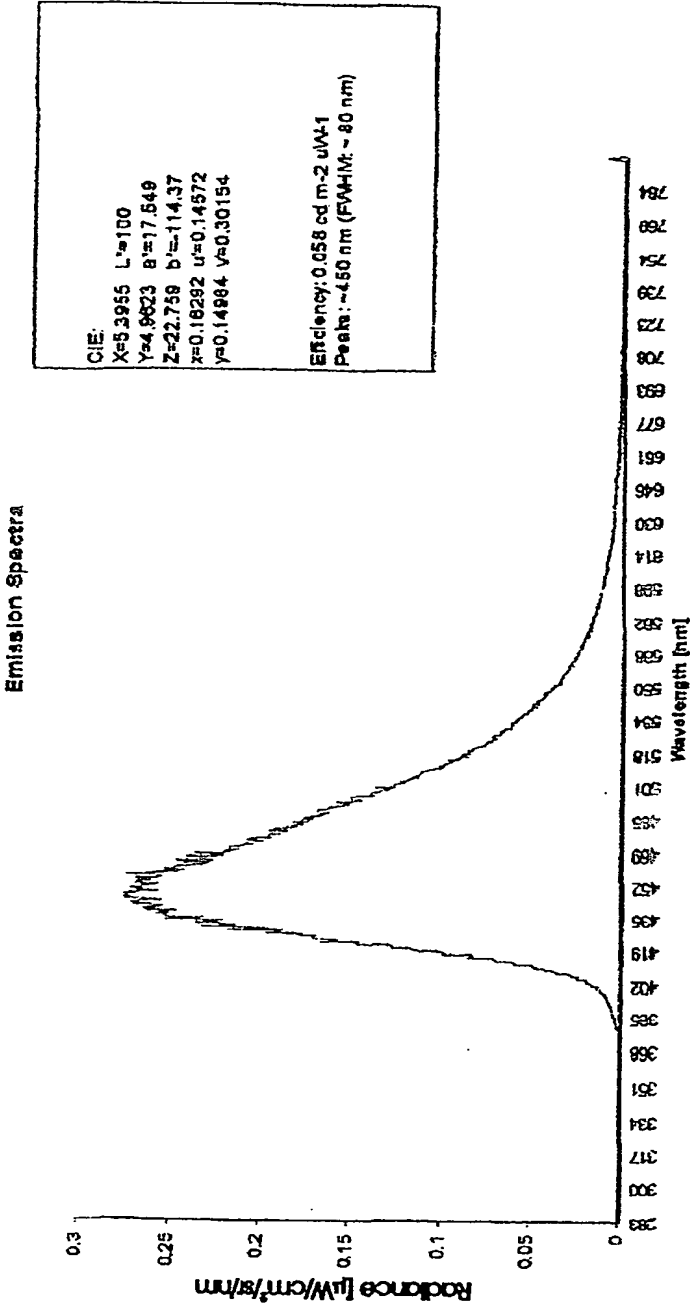


Fig 8

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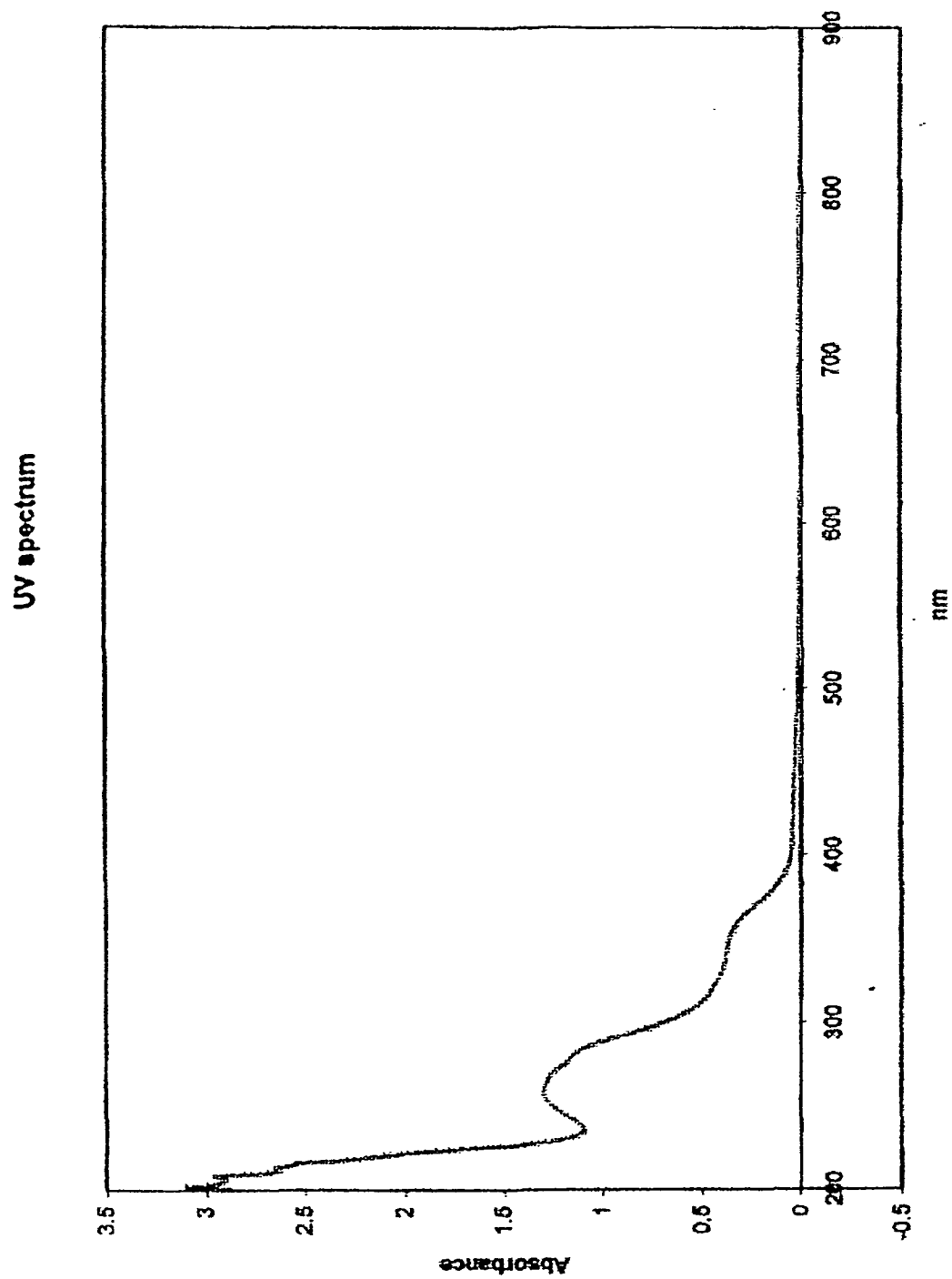


Fig 9

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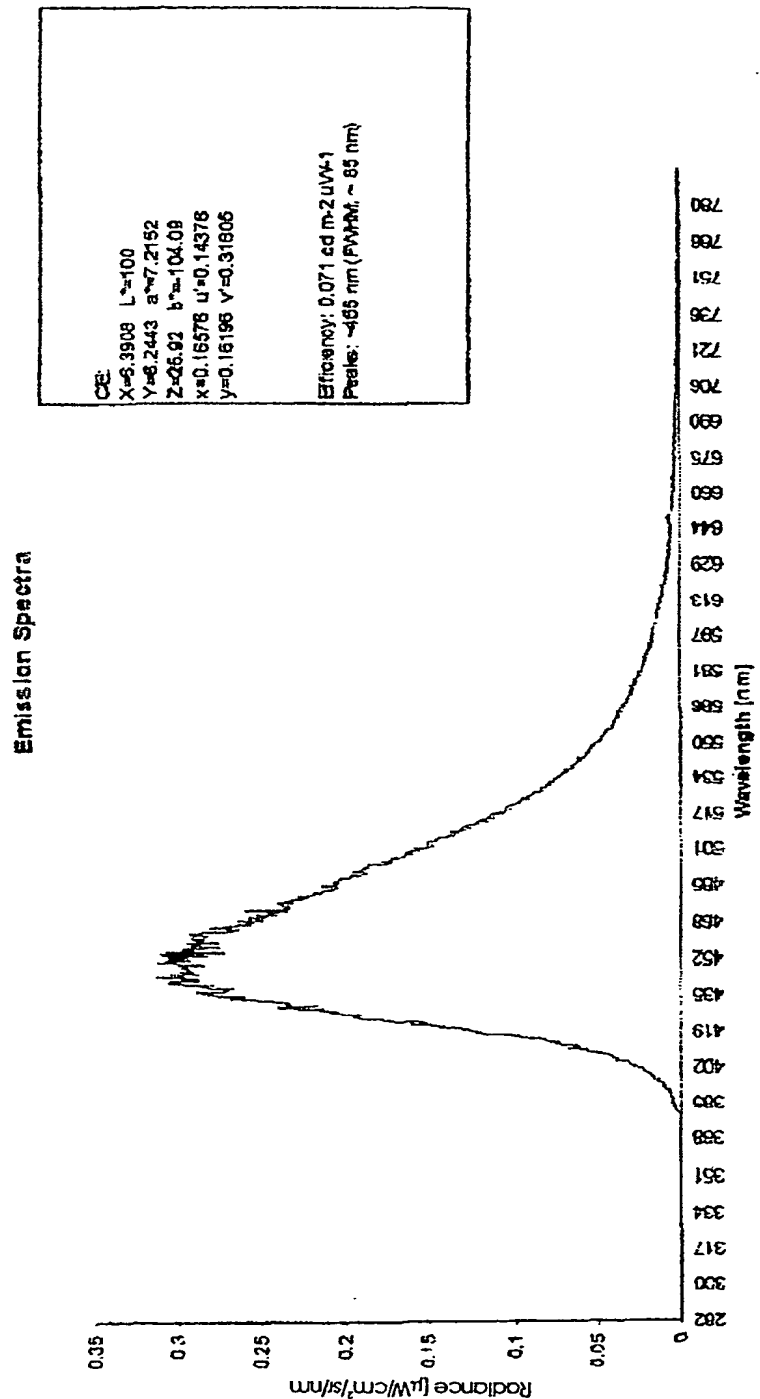


Fig 10

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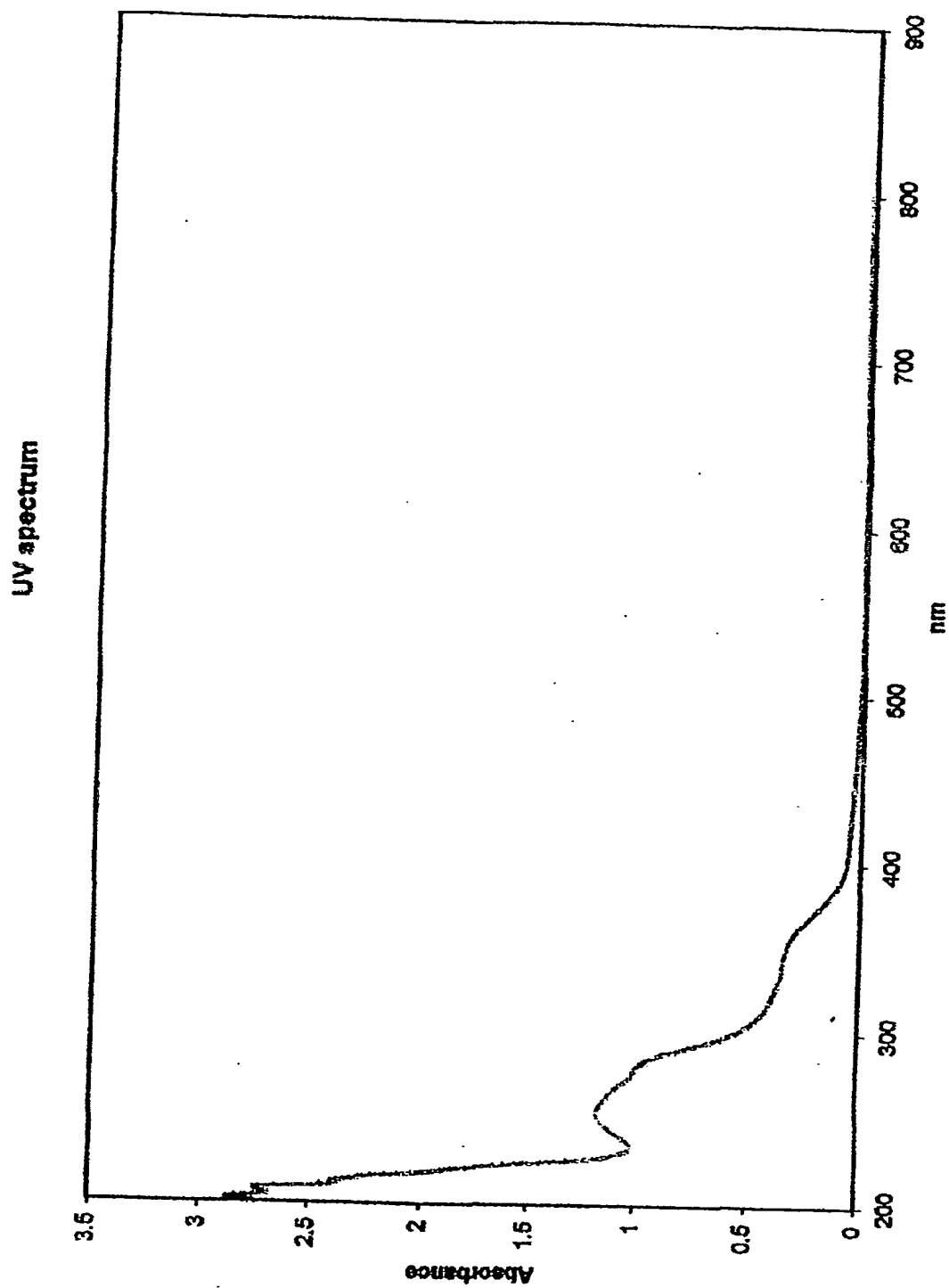


Fig 11



12/27

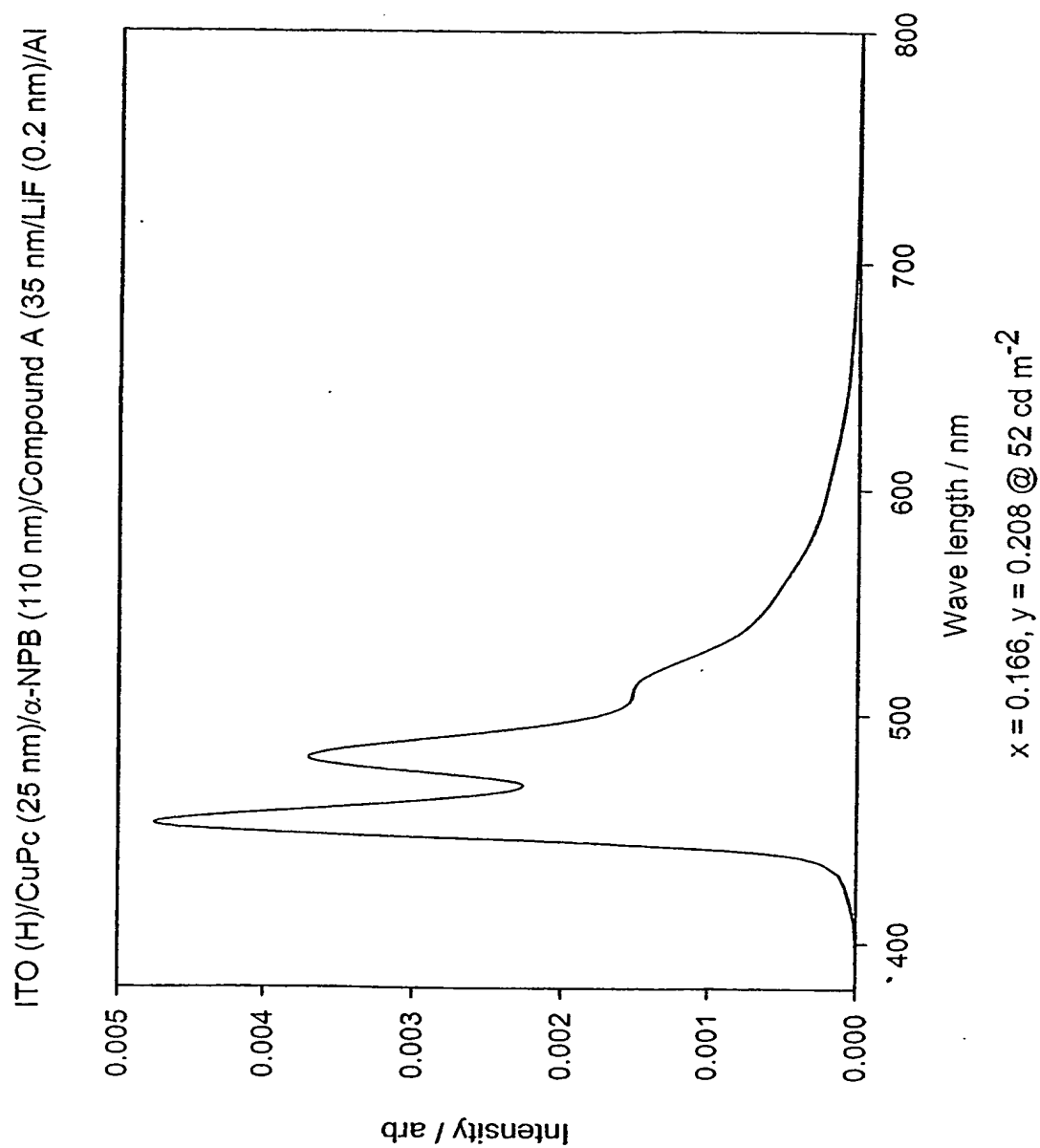


Fig 12

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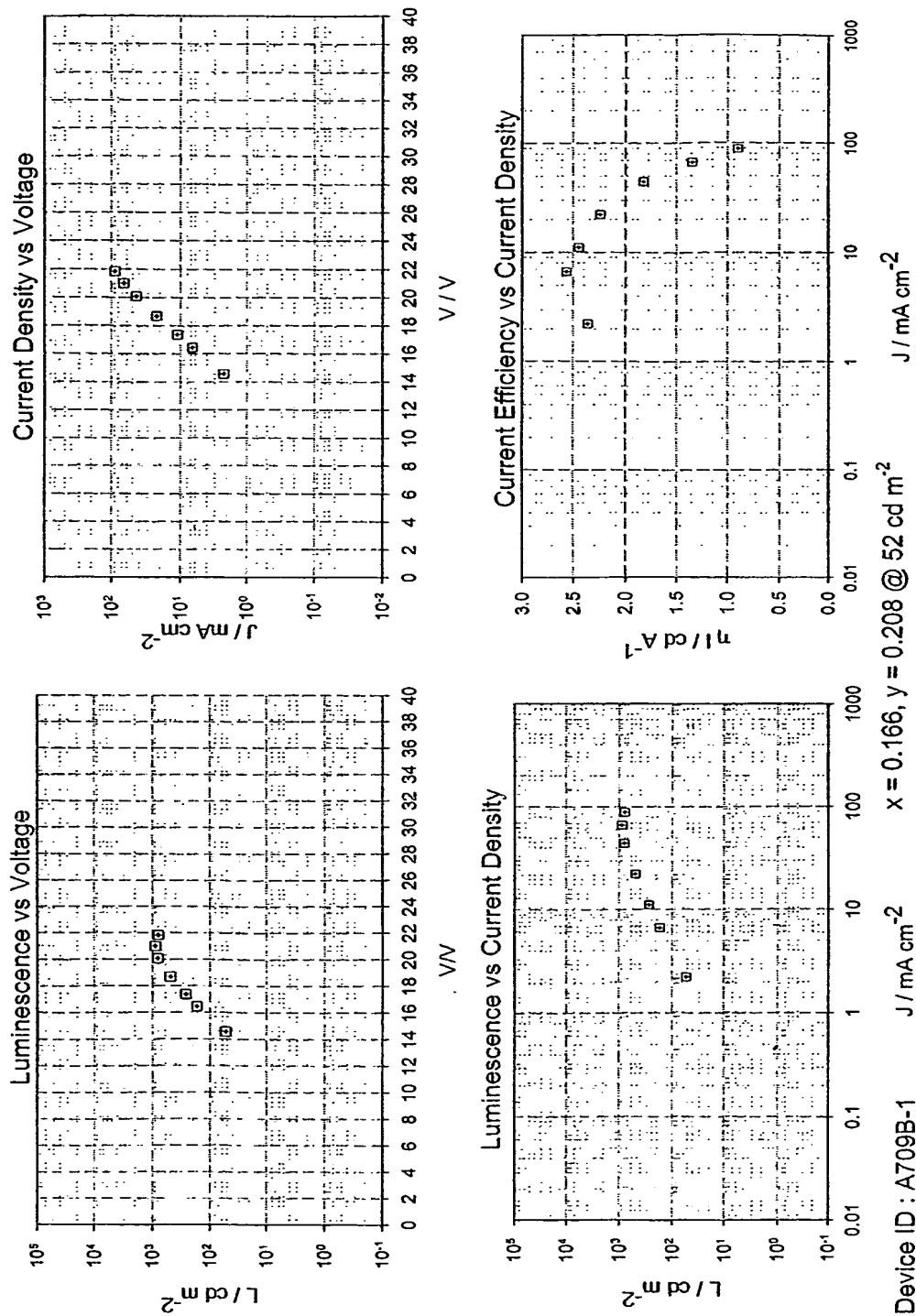
ITO (H)/CuPc (25 nm)/ $\alpha$ -NPB (110 nm)/Compound A (35 nm)/LiF (0.2 nm)/Al

Fig 13

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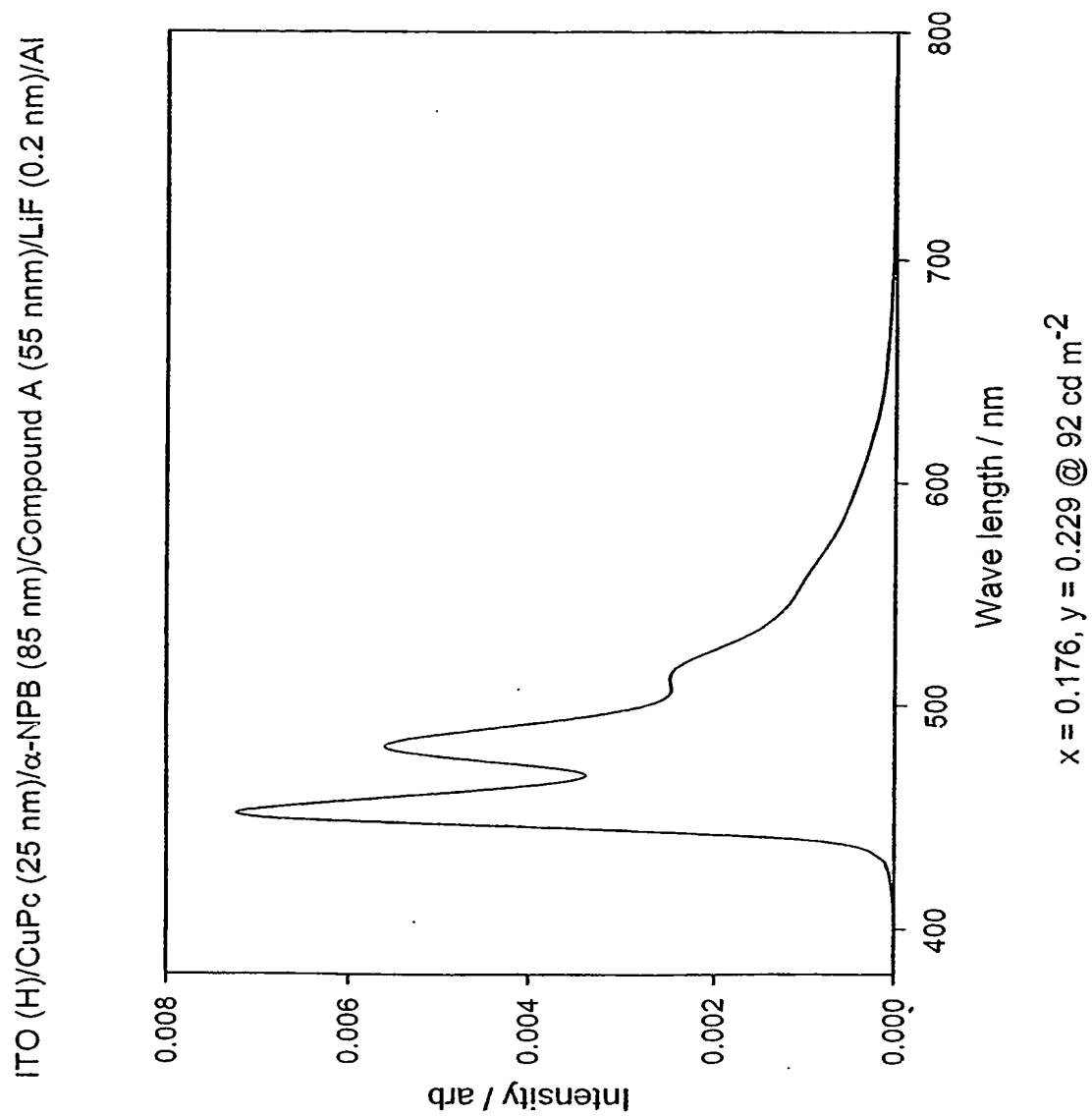


Fig 14

ITO (H)/CuPc (25 nm)/ $\alpha$ -NPB (85 nm)/Compound A (55 nm)/LiF (0.2 nm)/Al

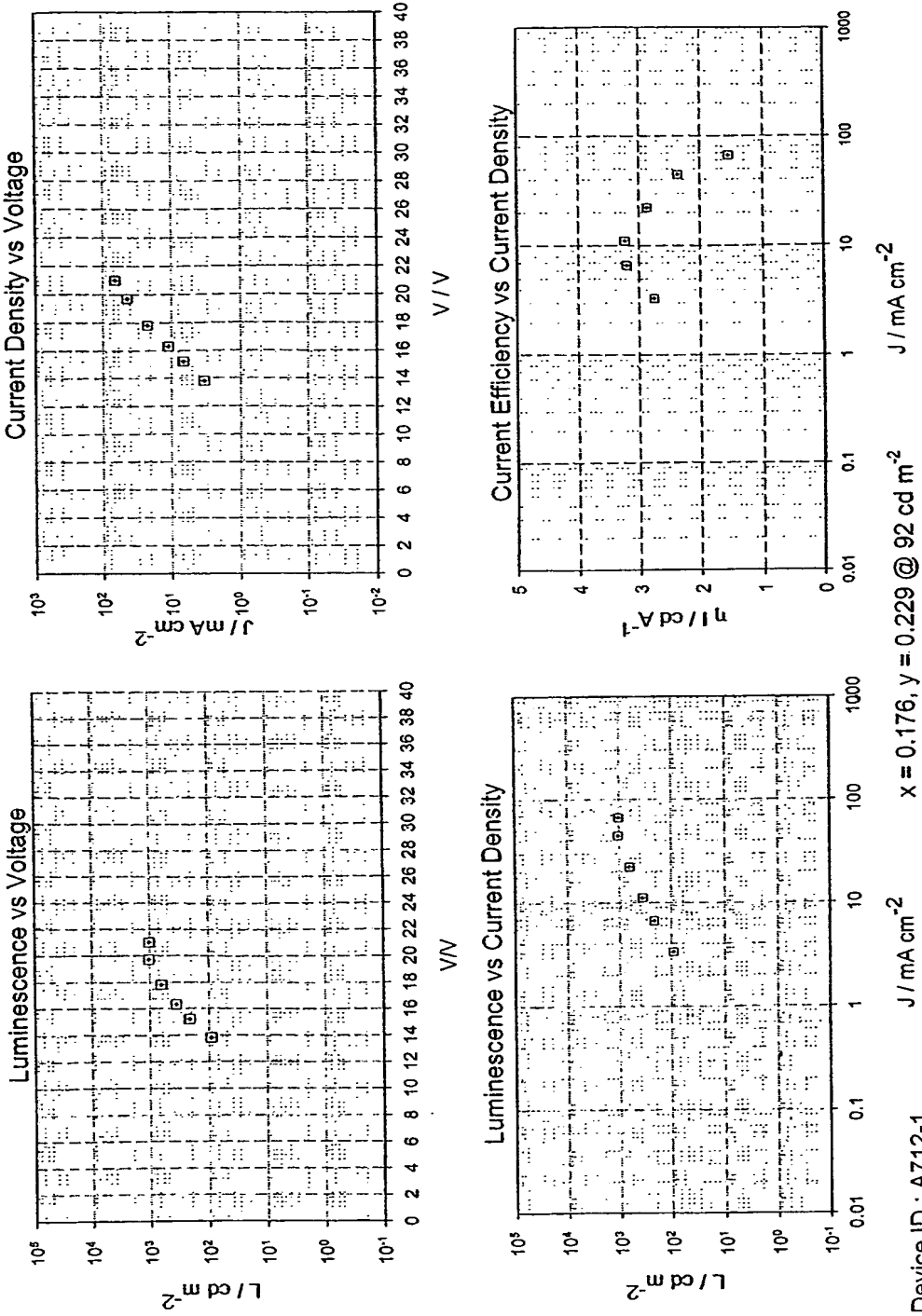


Fig 15

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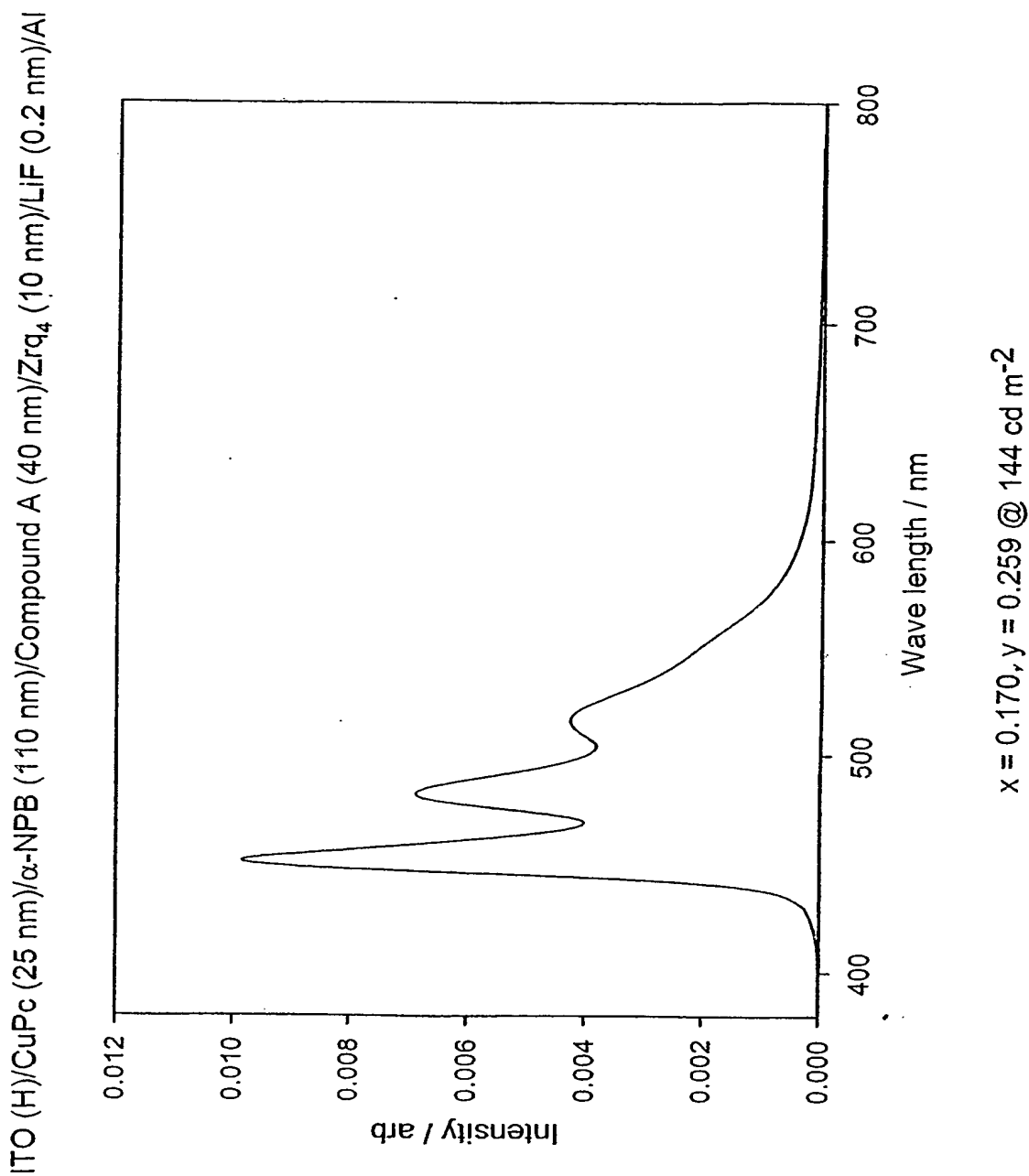


Fig 16

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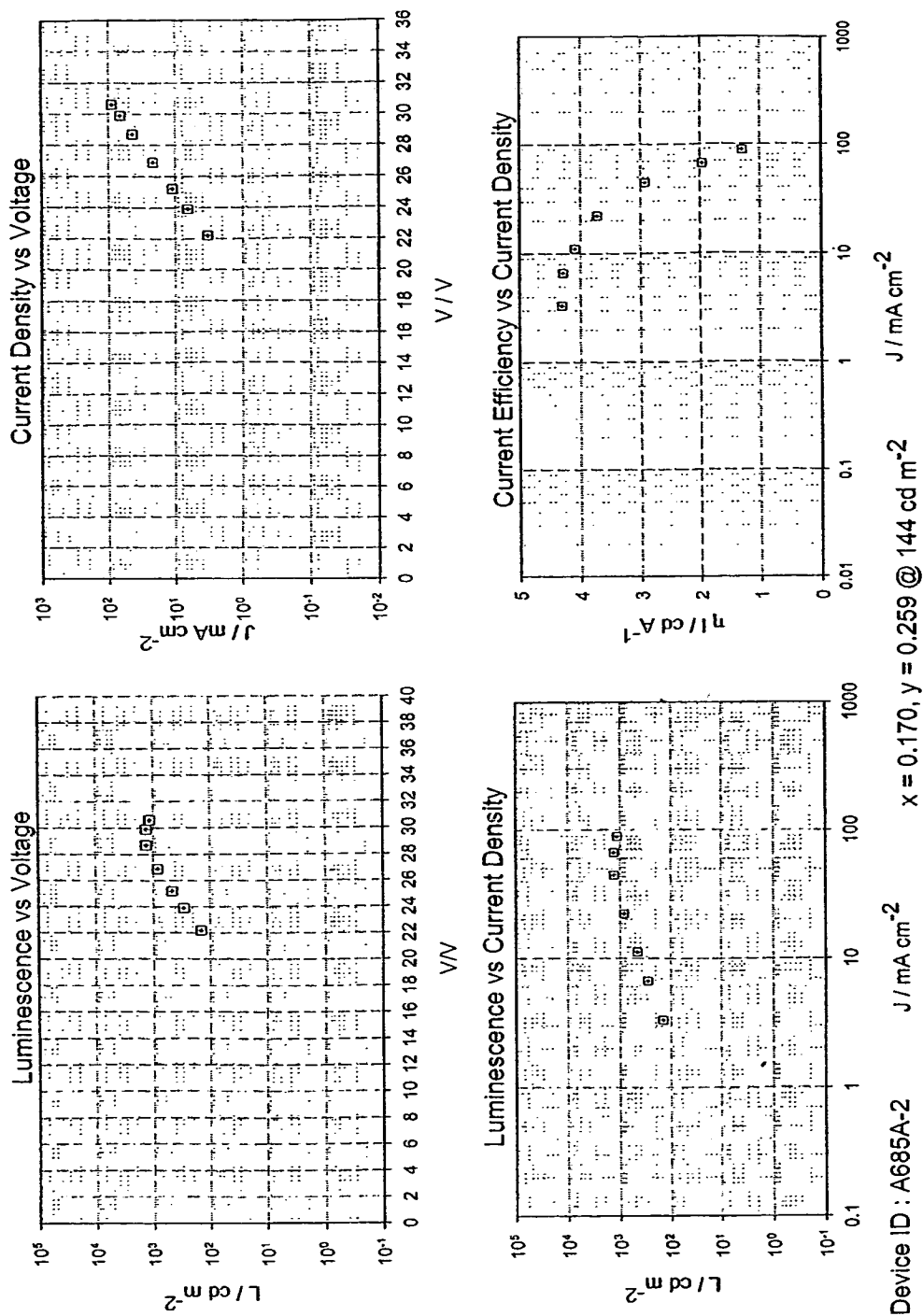
ITO (H)/CuPc (25 nm)/ $\alpha$ -NPB (110 nm)/Compound A (40 nm)/Zrqn (10 nm)/LiF (0.2 nm)/Al

Fig 17

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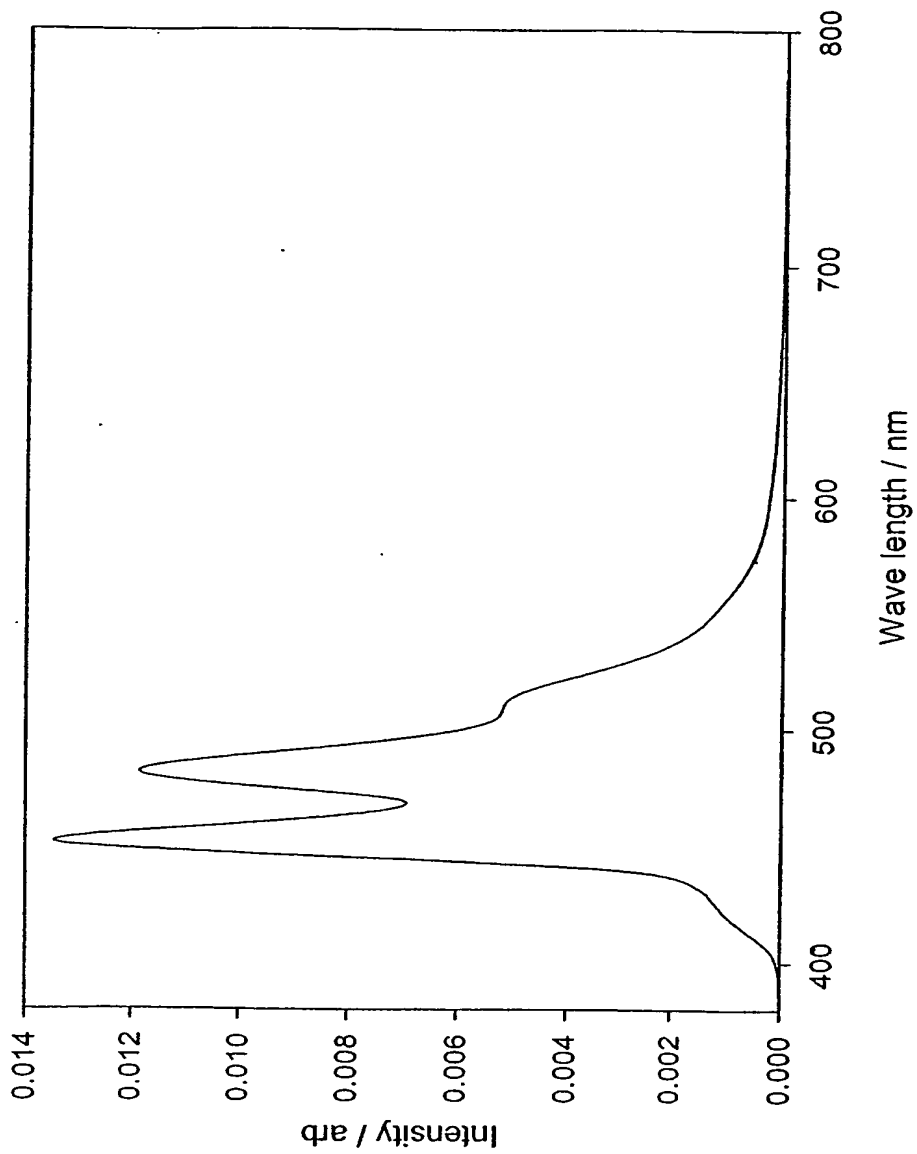
ITO (H)/CuPc (25 nm)/ $\alpha$ -NPB (110 nm)/Compound B (1 nm)/Compound A (25 nm)/Zrqr<sub>4</sub> (10 nm)/LiF (0.2 nm)/Alx = 0.145, y = 0.189 @ 141 cd m<sup>-2</sup>

Fig 18

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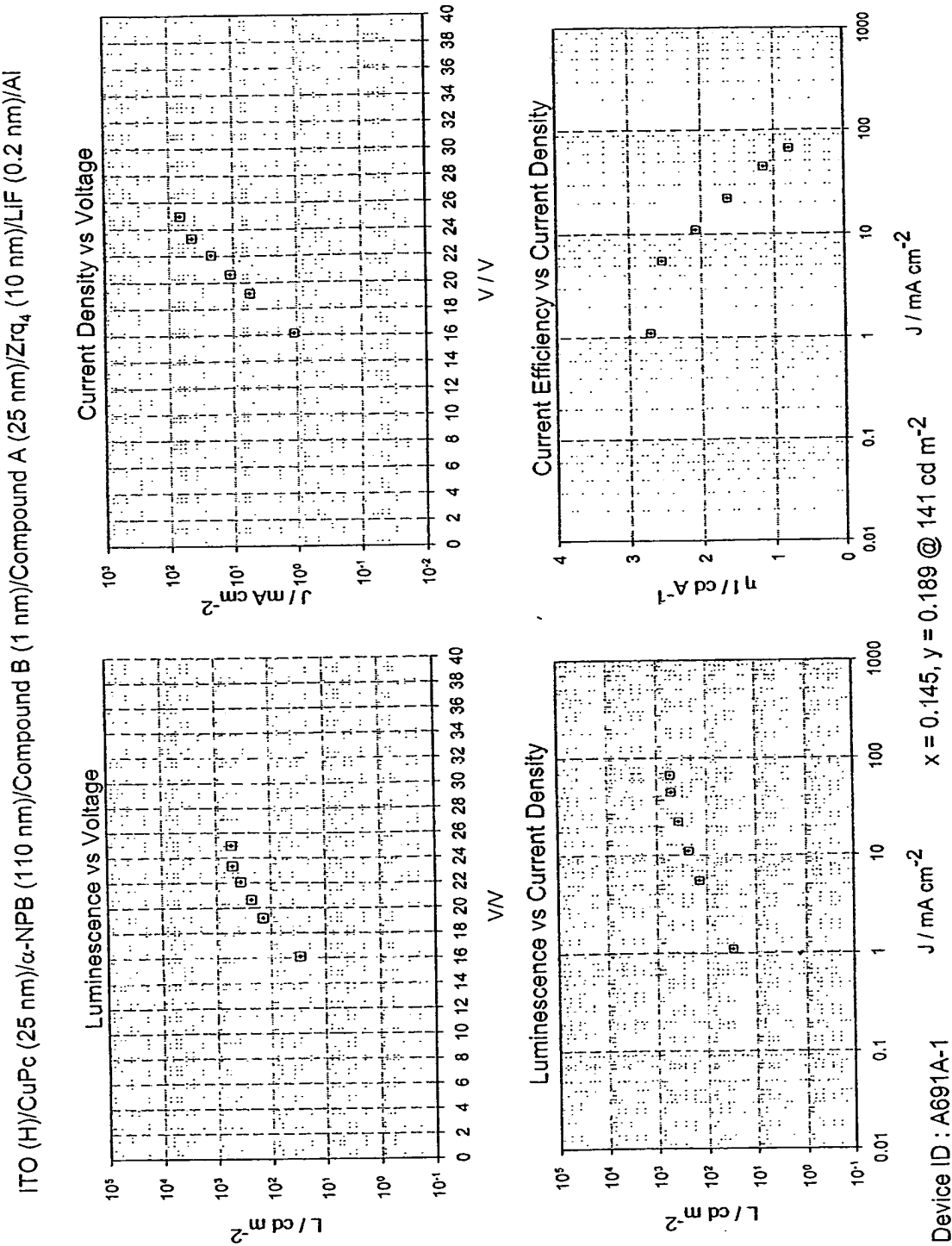


Fig 19



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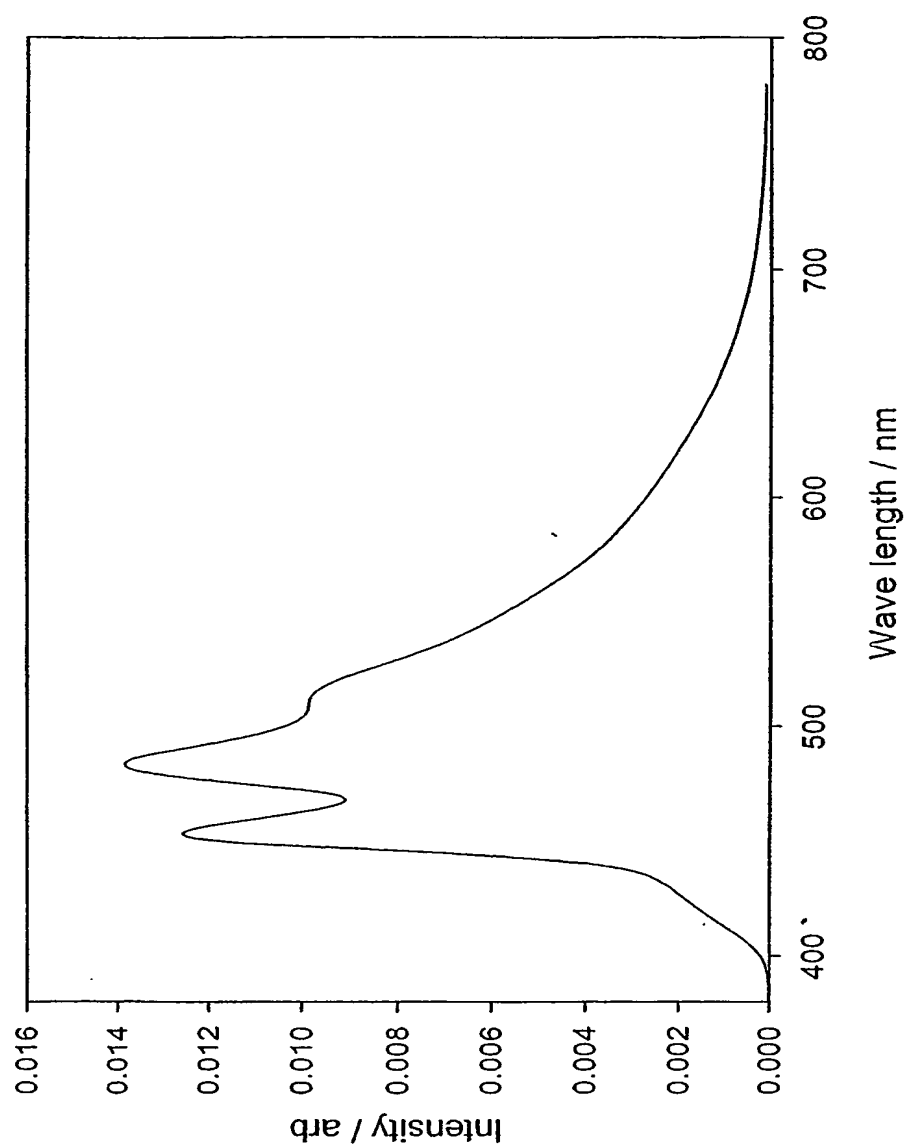
ITO (H)/CuPc (25 nm)/ $\alpha$ -NPB (110 nm)/Compound C (40 nm)/ZrO<sub>4</sub> (10 nm)/LiF (0.5 nm)/Al $x = 0.210, y = 0.290 @ 75 \text{ cd m}^{-2}$ 

Fig 20

ITO (H)/CuPc (25 nm)/ $\alpha$ -NPB (110 nm)/Compound C (40 nm)/ZrO<sub>4</sub> (10 nm)/LiF (0.5 nm)/Al

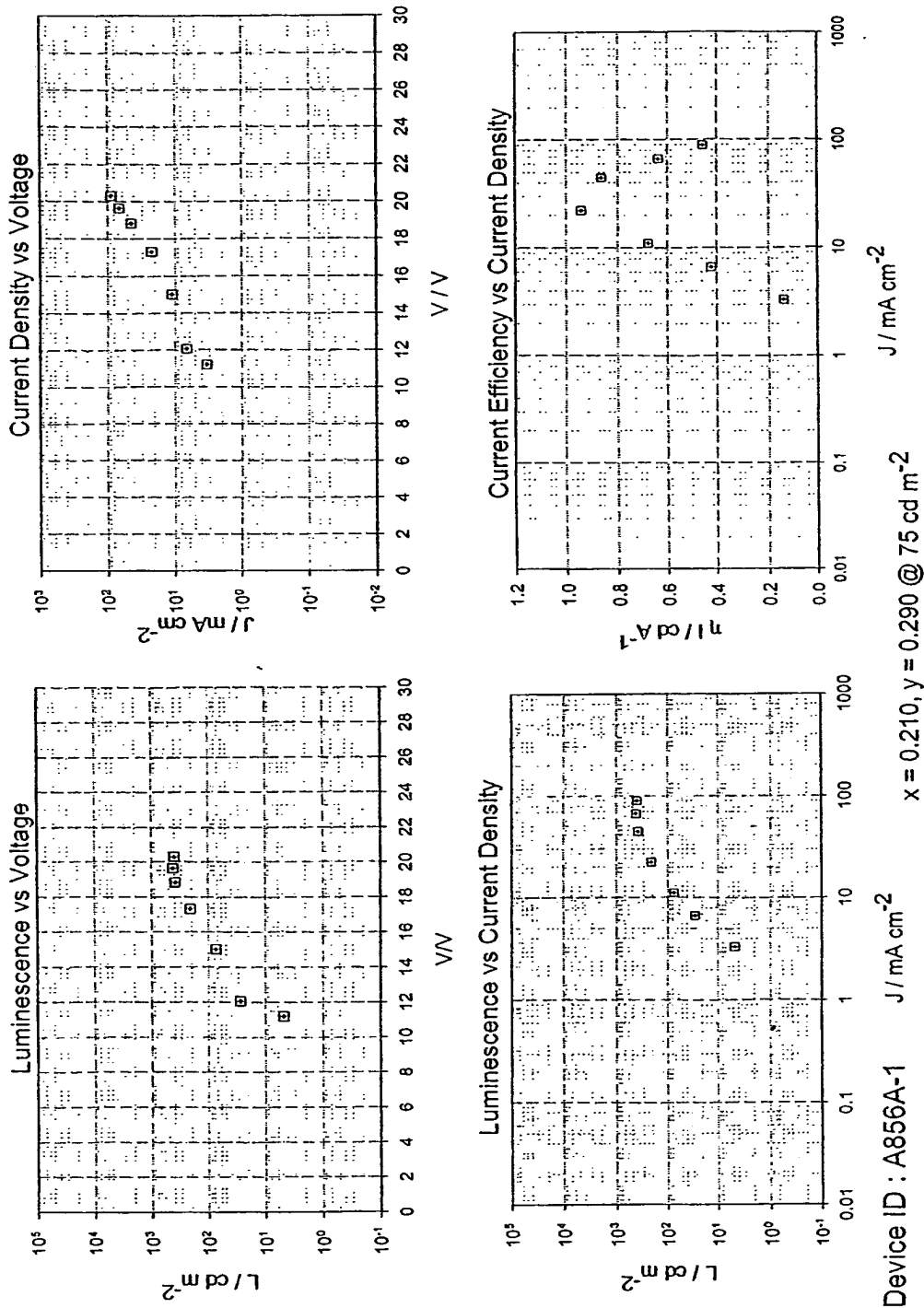


Fig 21

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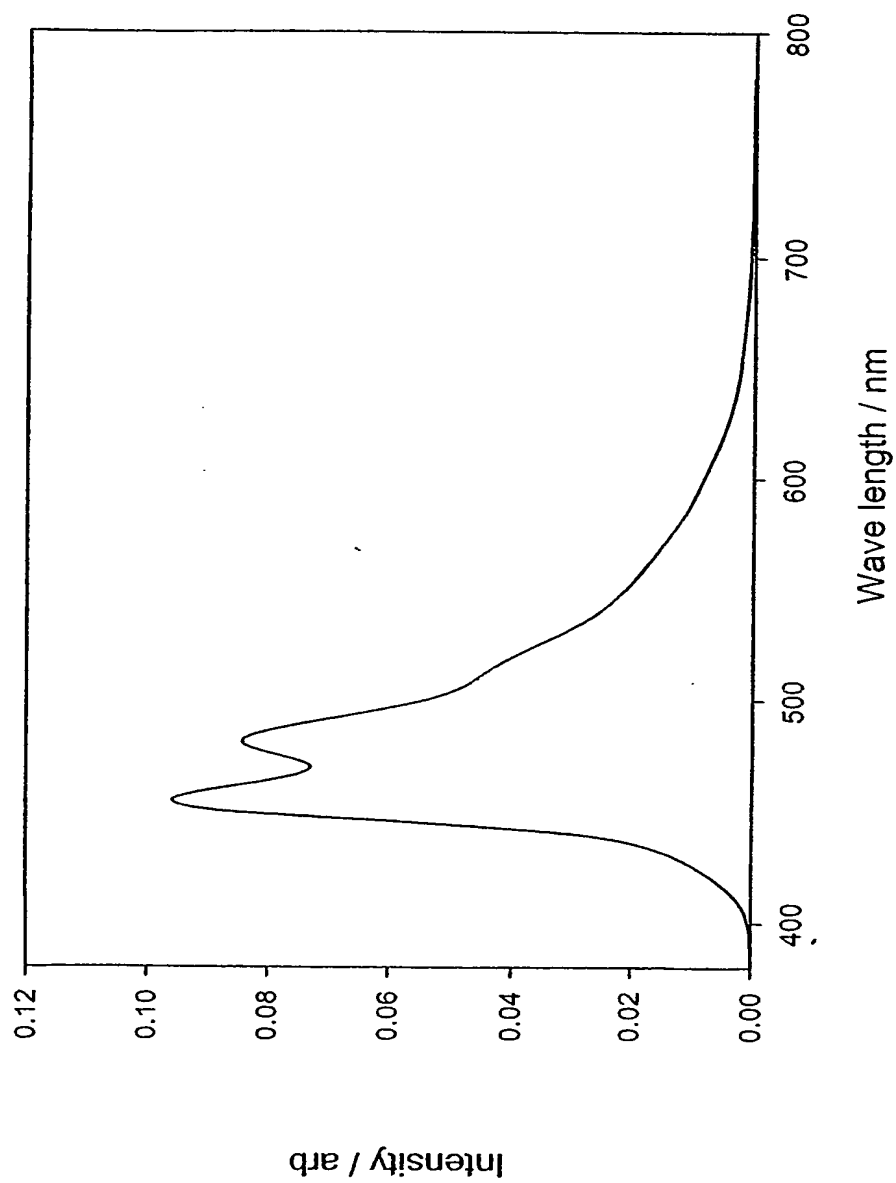
ITO (H)/CuPc (25 nm)/ $\alpha$ -NPB (75 nm)/BAIq<sub>2</sub> (15 nm)/Compound C (40 nm)/Zrqn<sub>4</sub> (10 nm)/LiF (0.5 nm)/Al $x = 0.171, y = 0.212 @ 74 \text{ cd m}^{-2}$ 

Fig 22

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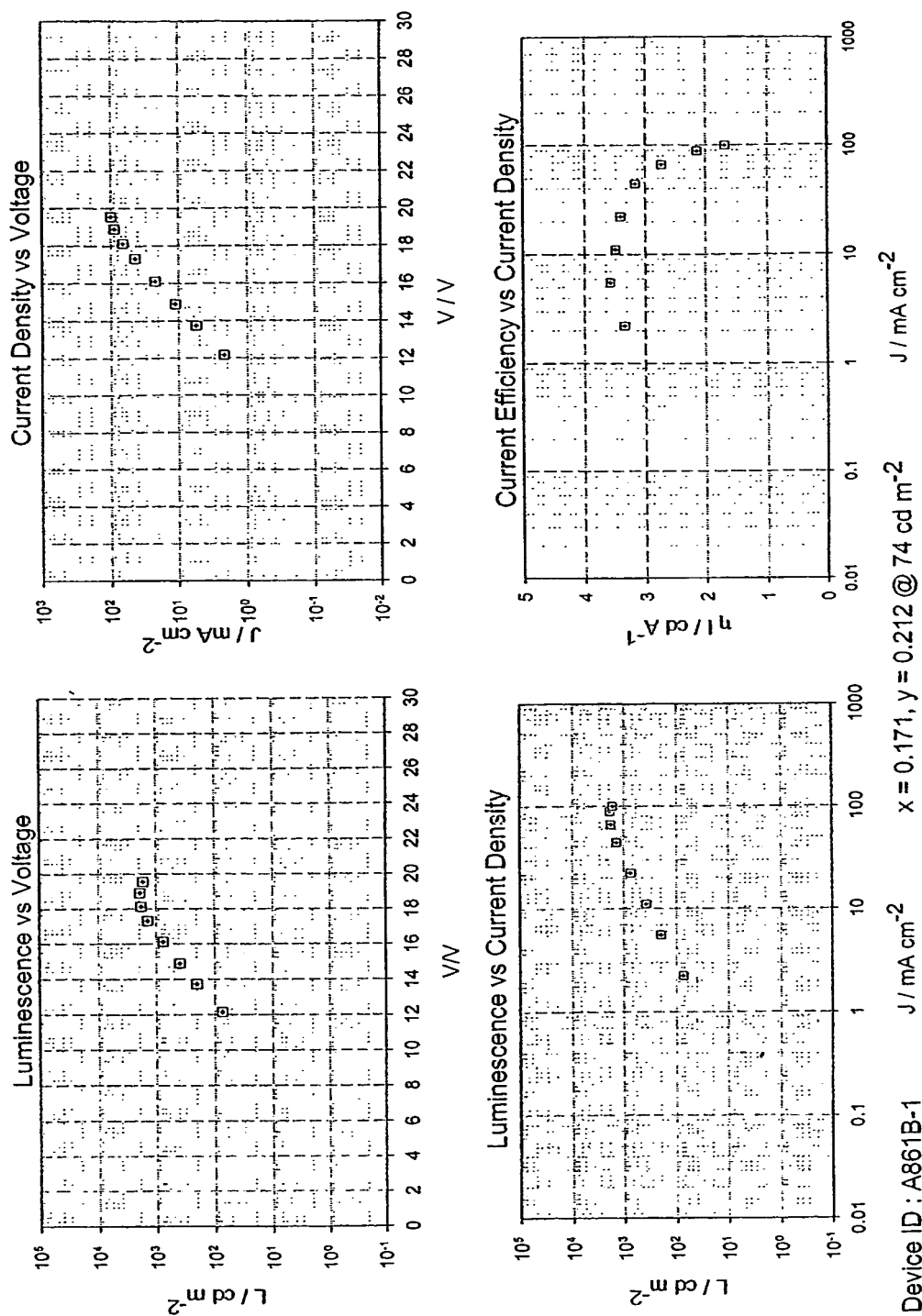
ITO (H)/CuPc (25 nm)/ $\alpha$ -NPB (75 nm)/BALq<sub>2</sub> (15 nm)/Compound C (40 nm)/Zrqn<sub>4</sub> (10 nm)/LiF (0.5 nm)/Al

Fig 23

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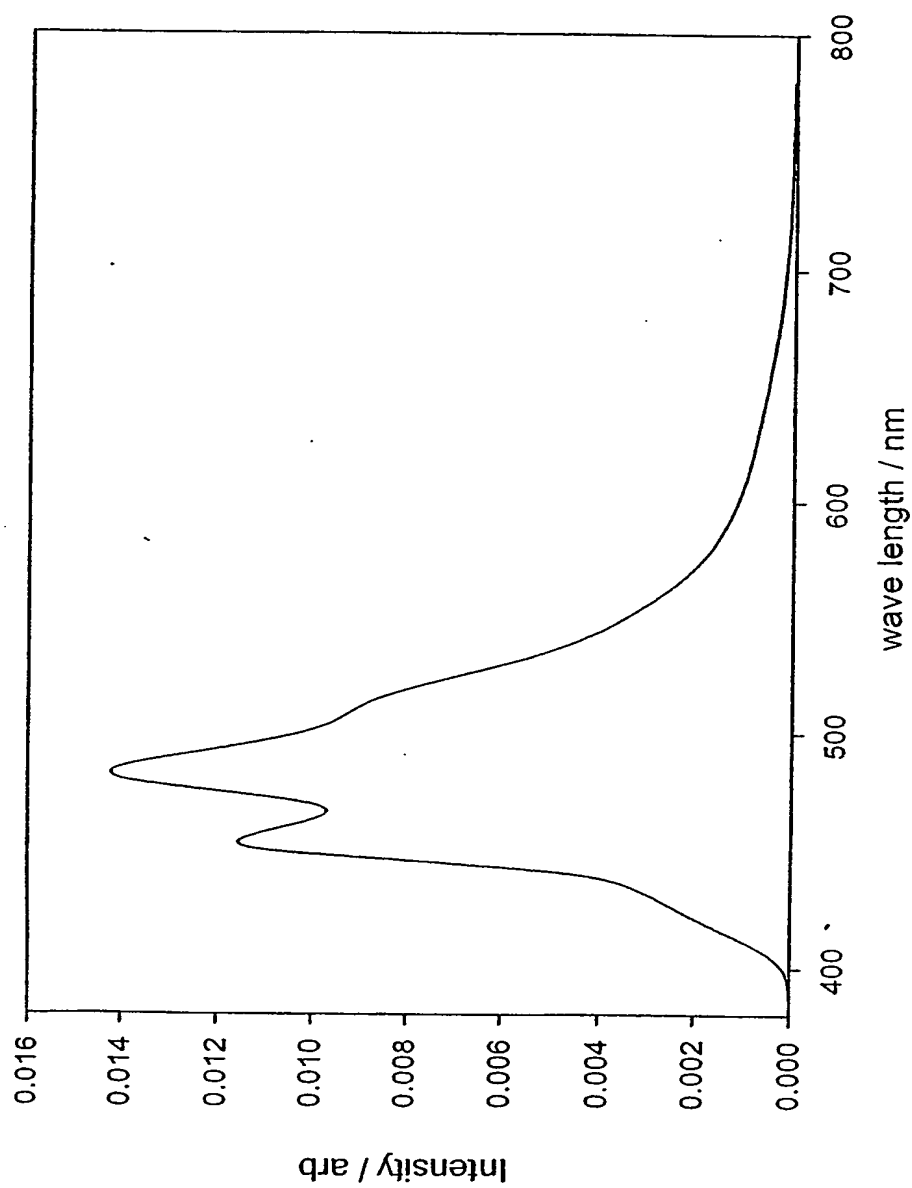
ITO (H)/CuPc (25 nm)/ $\alpha$ -NPB (110 nm)/Compound D (40 nm)/BAIq<sub>2</sub> (10 nm)/LiF (0.5 nm)/Al $x = 0.181, y = 0.290 @ 108 \text{ cd m}^{-2}$ 

Fig 24

ITO (H)/CuPc (25 nm)/ $\alpha$ -NPB (110 nm)/Compound D (40 nm)/BAIq<sub>2</sub> (10 nm)/LiF (0.5 nm)/Al

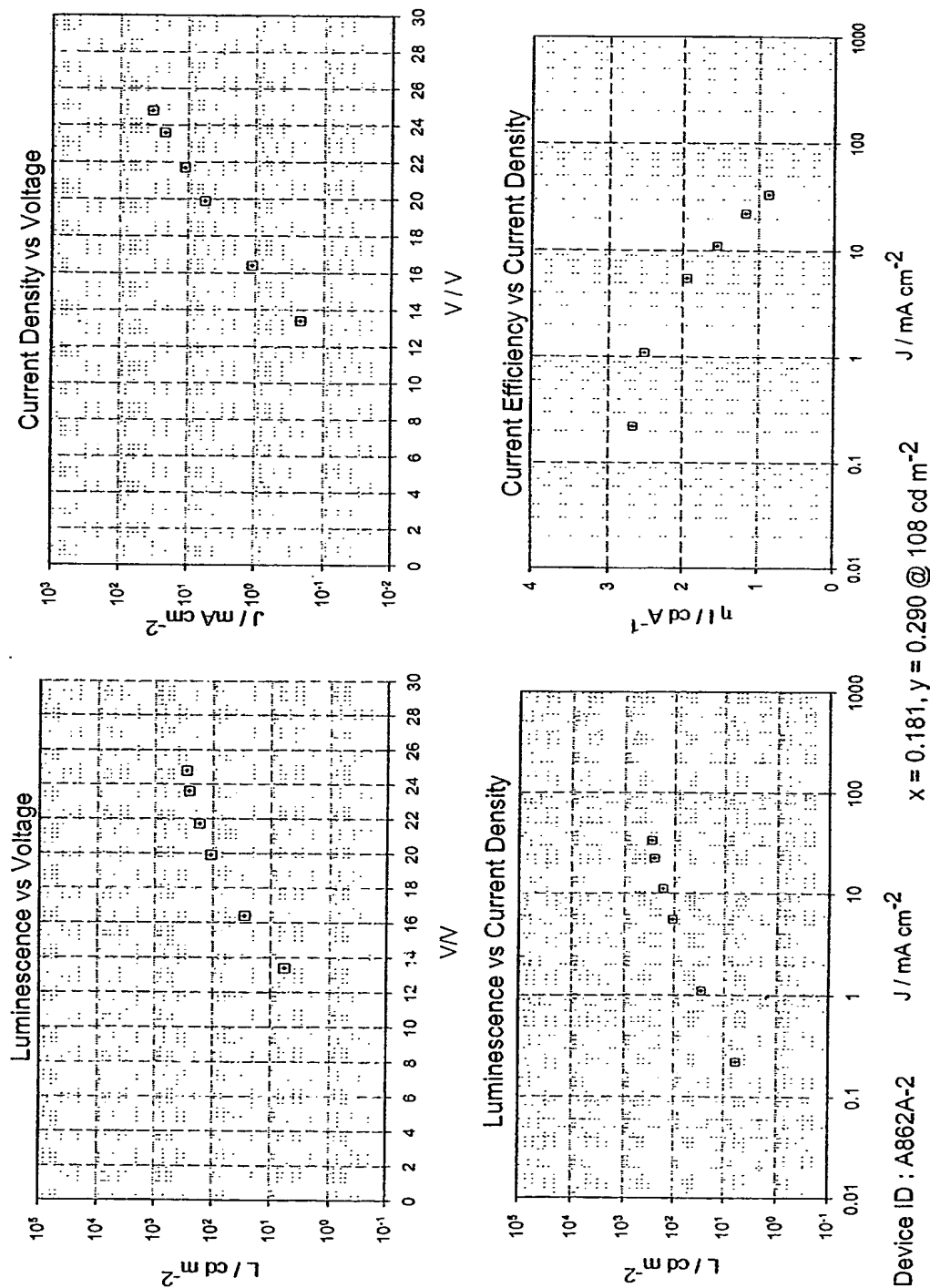
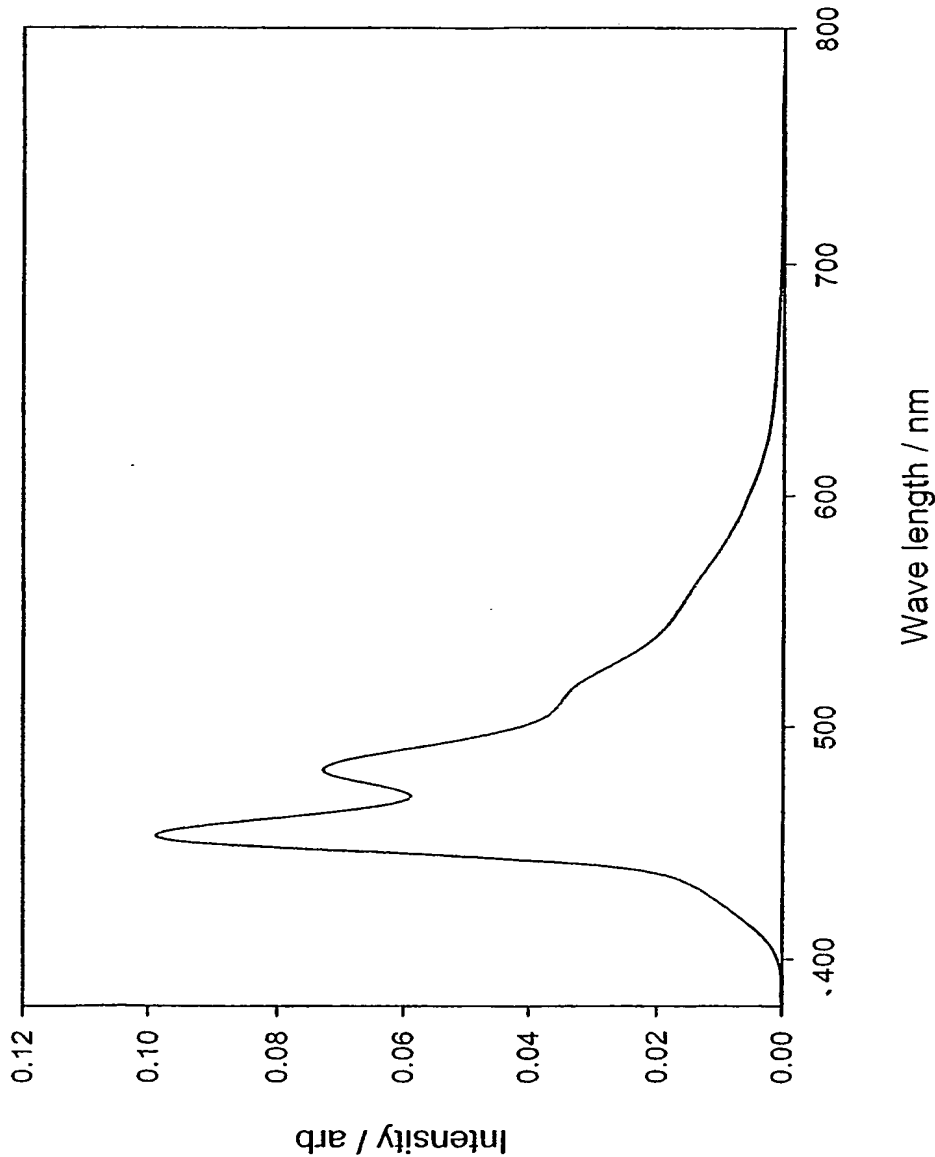


Fig 25

ITO (H)/CuPc (25 nm)/ $\alpha$ -NPB (60 nm)/Compound E:Perylene (30:0.02 nm)/LiF (0.5 nm)/Al



x = 0.178, y = 0.233 @ 163 cd m<sup>-2</sup>

Fig 26

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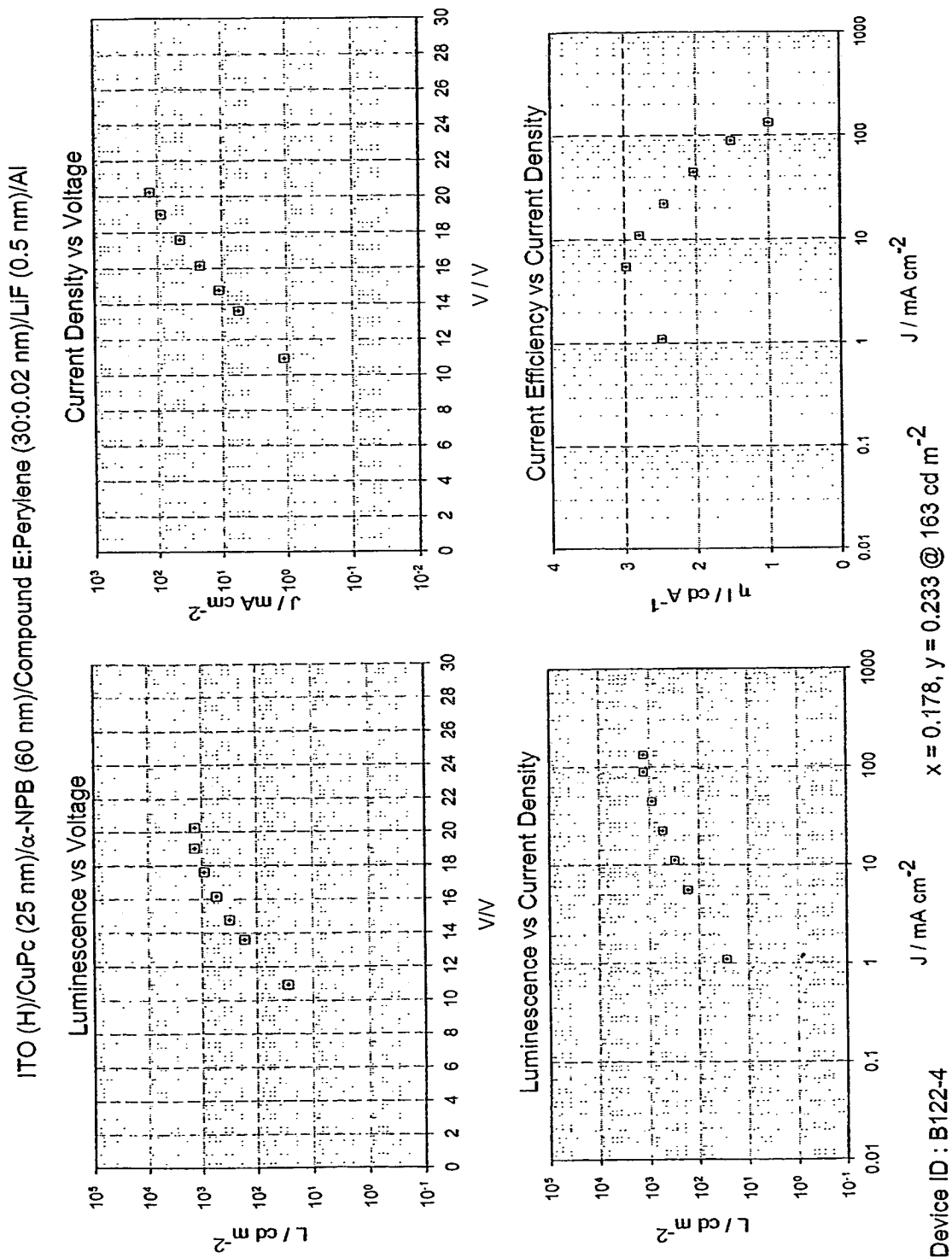


Fig 27



# INTERNATIONAL SEARCH REPORT

PCT/GB2004/001079

<b>A. CLASSIFICATION OF SUBJECT MATTER</b> IPC 7 H01L51/30 C09K11/06 H05B33/14 C07F5/02		
According to International Patent Classification (IPC) or to both national classification and IPC		
<b>B. FIELDS SEARCHED</b> Minimum documentation searched (classification system followed by classification symbols) IPC 7 H01L C09K H05B C07F		
Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched		
Electronic data base consulted during the international search (name of data base and, where practical, search terms used) EPO-Internal, CHEM ABS Data		
<b>C. DOCUMENTS CONSIDERED TO BE RELEVANT</b>		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
P, X	CHENG, CHUNG-CHIH ET AL: "Syntheses and remarkable photophysical properties of 5-(2-pyridyl) pyrazolate boron complexes; photoinduced electron transfer" CHEMICAL COMMUNICATIONS, no. 20, 3 October 2003 (2003-10-03), pages 2628-2629, XP002286359 ISSN: 1359-7345 compound 2d	1-4, 6, 7, 10, 12-31, 35
A	US 6 368 731 B1 (HEUER HELMUT-WERNER ET AL) 9 April 2002 (2002-04-09) cited in the application the whole document	1-36
-/--		
<input checked="" type="checkbox"/> Further documents are listed in the continuation of box C.		
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Date of the actual completion of the international search 29 June 2004		Date of mailing of the international search report 12/07/2004
Name and mailing address of the ISA European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016		Authorized officer Elliott, A

## INTERNATIONAL SEARCH REPORT

PCT/GB2004/001079

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	LIU Y ET AL: "Highly efficient white organic electroluminescence from a double-layer device based on a boron hydroxyphenylpyridine complex" ANGEWANDTE CHEMIE, INTERNATIONAL EDITION, vol. 41, no. 1, 4 January 2002 (2002-01-04), pages 182-184, XP002286360 ISSN: 1433-7851 the whole document	1-36
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A	LI ET AL: "A mixed pyridine-phenol boron complex as an organic electroluminescent material" CHEMICAL COMMUNICATIONS, no. 16, 2000, pages 1551-1552, XP002231558 ISSN: 1359-7345 the whole document	1-36
A	WU Q ET AL: "Synthesis, structure and electroluminescence of BR2q (R = Et, Ph, 2-naphthyl and q = 8-hydroxyquinolato)" CHEMISTRY OF MATERIALS, vol. 12, no. 1, January 2000 (2000-01), pages 79-83, XP000919563 ISSN: 0897-4756 the whole document	1-36

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PCT/GB2004/001079

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